

---

## Chapter 2: Changes in Atmospheric Constituents and in Radiative Forcing

**Coordinating Lead Authors:** Piers Forster (UK), V. Ramaswamy (USA)

**Lead Authors:** Paulo Artaxo (Brasil), Terje Berntsen (Norway), Richard A. Betts (UK), David W. Fahey (USA), James Haywood (UK), Judith Lean (USA), David C. Lowe (New Zealand), Gunnar Myhre (Norway), John Nganga (Kenya), Ronald Prinn (USA, New Zealand), Graciela Raga (Mexico, Argentina), Michael Schulz (France, Germany), Robert Van Dorland (Netherlands)

**Contributing Authors:** Greg Bodeker (New Zealand), Olivier Boucher (UK, France), William Collins (USA), Thomas J. Conway (USA), Ed Dlugokencky (USA), James W. Elkins (USA), David Etheridge (Australia), Peter Foukal (USA), Paul Fraser (Australia), Marvin Geller (USA), Fortunat Joos (Switzerland), C. David Keeling (USA), Ralph Keeling (USA), Stefan Kinne (Germany), Keith Lassey (New Zealand), Ulrike Lohmann (Switzerland), Andrew Manning (UK, New Zealand), Steve Montzka (USA), David Oram (UK), Kath O'Shaughnessy (New Zealand), Stephen Piper (USA), Gian-Kasper Plattner (Switzerland), Michael Ponater (Germany), Navin Ramankutty (USA, India), George Reid (USA), David Rind (USA), Karen Rosenlof (USA), Robert Sausen (Germany), Dan Schwarzkopf (USA), S. Solanki (Germany), Georgiy Stenchikov (USA), Nicola Stuber (UK, Germany), Toshihiko Takemura (Japan), Christiane Textor (France, Germany), Ray Wang (USA), Ray Weiss (USA), Tim Whorf (USA)

**Review Editors:** Teruyuki Nakajima (Japan), V. Ramanathan (USA)

**Date of Draft:** 27 October 2006

---

1	<b>Table of Contents</b>	
2		
3	Executive Summary.....	3
4	2.1 Introduction and Scope .....	6
5	2.2 Concept of Radiative Forcing (RF).....	6
6	2.3 Chemically and Radiatively Important Gases.....	8
7	2.3.1 Atmospheric Carbon Dioxide (CO <sub>2</sub> ).....	8
8	2.3.2 Atmospheric Methane (CH <sub>4</sub> ).....	11
9	2.3.3 Other Kyoto Protocol Gases (N <sub>2</sub> O, HFCs, PFCs, and SF <sub>6</sub> ).....	13
10	2.3.4 Montreal Protocol Gases (CFCs and HCFCs).....	15
11	2.3.5 Trends in the Hydroxyl Free Radical (OH).....	16
12	2.3.6 Ozone.....	18
13	2.3.7 Stratospheric Water Vapour.....	21
14	2.3.8 Observations of Long-Lived Greenhouse Gas Radiative Effects.....	22
15	2.4 Aerosols.....	22
16	2.4.1 Introduction and Summary of the Third Assessment Report.....	22
17	2.4.2 Developments Related to Aerosol Observations.....	24
18	2.4.3 Advances in Modelling the Aerosol Direct Effect.....	27
19	2.4.4 Estimates of Aerosol Direct Radiative Forcing.....	28
20	2.4.5 Aerosol Influence on Clouds (Cloud Albedo Effect).....	37
21	2.5 Anthropogenic Changes in Surface Albedo and the Surface Energy Budget.....	44
22	2.5.1 Introduction.....	44
23	2.5.2 Changes in Land Cover Since 1750.....	45
24	2.5.3 Radiative Forcing by Anthropogenic Surface Albedo Change: Land-use.....	45
25	2.5.4 Radiative Forcing by Anthropogenic Surface Albedo Change: Black Carbon in Snow and Ice.....	47
26	2.5.5 Other Effects of Anthropogenic Changes in Land Cover.....	47
27	2.5.6 Tropospheric Water Vapour from Anthropogenic Sources.....	48
28	2.5.7 Anthropogenic Heat Release.....	48
29	2.5.8 Effects of CO <sub>2</sub> Changes on Climate via Plant Physiology: “Physiological Forcing”.....	48
30	2.6 Contrails and Aircraft-Induced Cloudiness.....	49
31	2.6.1 Introduction.....	49
32	2.6.2 Radiative Forcing Estimates for Persistent Line-Shaped Contrails.....	49
33	2.6.3 Radiative Forcing Estimates for Aviation-Induced Cloudiness.....	49
34	2.6.4 Aviation Aerosols.....	51
35	2.7 Natural Forcings.....	51
36	2.7.1 Solar Variability.....	51
37	2.7.2 Explosive Volcanic Activity.....	56
38	2.8 Utility of Radiative Forcing.....	58
39	2.8.1 Vertical Forcing Patterns and Surface Energy Balance Changes.....	59
40	2.8.2 Spatial Patterns of Radiative Forcing.....	59
41	2.8.3 Alternative Methods of Calculating Radiative Forcing.....	59
42	2.8.4 Linearity of the Forcing-Response Relationship.....	60
43	2.8.5 Efficacy and Effective Radiative Forcing.....	60
44	2.8.6 Efficacy and the Forcing-Response Relationship.....	62
45	2.9 Synthesis.....	63
46	2.9.1 Uncertainties in Radiative Forcing.....	63
47	2.9.2 Global Mean Radiative Forcing.....	64
48	2.9.3 Global-Mean Radiative Forcing by Emission Precursor.....	65
49	2.9.4 Future Climate Impact of Current Emissions.....	65
50	2.9.5 Time Evolution of Radiative Forcing and Surface Forcing.....	66
51	2.9.6 Spatial Patterns of Radiative Forcing and Surface Forcing.....	67
52	2.10 GWPs and Other Metrics for Comparing Different Emissions.....	67
53	2.10.1 Definition of an emission Metric and the GWP.....	67
54	2.10.2 Direct GWPs.....	69
55	2.10.3 Indirect GWPs.....	70
56	2.10.4 New Alternative Metrics for Assessing Emissions.....	71
57	References.....	73
58	Frequently Asked Question 2.1: How Do Human Activities Contribute to Climate Change and How Do They Compare	
59	With Natural Influences?.....	104
60	Tables.....	107

## 1 Executive Summary

2  
3 Radiative forcing (RF)<sup>1</sup> is a concept used for quantitative comparisons of the strength of different human and  
4 natural agents in causing climate change. Climate model studies since the IPCC WG1 Third Assessment  
5 Report (IPCC, 2001) (hereinafter TAR) give medium confidence that the equilibrium global mean  
6 temperature response to a given RF is approximately the same (to within 25%) for most drivers of climate  
7 change.

8  
9 For the first time, the combined RF for all anthropogenic agents is derived. Estimates are also made for the  
10 first time of the separate RF components associated with the emissions of each agent.

11  
12 The combined anthropogenic RF is estimated to be  $+1.6 [-1.0, +0.8]^2 \text{ W m}^{-2}$ , indicating that, since 1750, it is  
13 *extremely likely*<sup>3</sup> that humans have exerted a substantial warming influence on climate. This RF estimate is  
14 *likely* to be at least 5 times greater than that due to solar irradiance changes. For the period 1950-2005 it is  
15 *exceptionally unlikely* that the combined natural RF (solar irradiance+ volcanic aerosol) has had a warming  
16 influence comparable to that of the combined anthropogenic RF.

17  
18 Increasing concentrations of the long-lived greenhouse gases (CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, halocarbons and SF<sub>6</sub>  
19 hereinafter LLGHGs) have led to a combined RF of  $+2.63 [\pm 0.26] \text{ W m}^{-2}$ . Their RF has a *high* level of  
20 scientific understanding<sup>4</sup>. The 9% increase in this RF since the TAR is the result of concentration changes  
21 since 1998.

- 22  
23 • The global mean concentration of carbon dioxide (CO<sub>2</sub>) in 2005 was 379 ppm, leading to an RF of  
24  $+1.66 [\pm 0.17] \text{ W m}^{-2}$ . Past emissions of fossil fuels and cement production have *likely* contributed  
25 about three quarters of the current RF, with the remainder caused by land-use changes. For the 1995–  
26 2005 decade the growth rate of CO<sub>2</sub> in the atmosphere was  $1.9 \text{ ppm yr}^{-1}$  and the CO<sub>2</sub> RF increased by  
27 20%, this is the largest change observed or inferred for any decade in at least the last 200 years. From  
28 1999 to 2005 global emissions from fossil fuel and cement production have been increasing at a rate  
29 of roughly  $3\% \text{ year}^{-1}$ .
  - 30  
31 • The global mean concentration of methane (CH<sub>4</sub>) in 2005 was 1774 ppb, contributing an RF of  $+0.48$   
32  $[\pm 0.05] \text{ W m}^{-2}$ . Over the past two decades methane growth rates in the atmosphere have generally  
33 decreased. The cause of this is not well understood. However, this decrease and the negligible long-  
34 term change in its main sink (OH), imply that total methane emissions are not increasing.
  - 35  
36 • The Montreal Protocol gases (CFCs, HCFCs, and chlorocarbons) as a group contributed  $+0.32$   
37  $[\pm 0.03] \text{ W m}^{-2}$  to the RF in 2005. Their RF peaked in 2003 and is now beginning to decline.
- 38

---

<sup>1</sup> The RF represents the stratospherically adjusted radiative flux change evaluated at the tropopause, as defined in the IPCC WG1 Third Assessment Report (IPCC, 2001). Positive RFs lead to a global mean surface warming and negative RFs to a global mean surface cooling. RF is not, however, designed as an indicator of the detailed aspects of climate response. Unless otherwise mentioned, RF here refers to global-mean radiative forcing. RFs are calculated in various ways depending on the agent: from changes in emissions and/or changes in concentrations, and from observations and other knowledge of climate change drivers. In this Report, the RF value for each agent is reported as the difference in RF, unless otherwise mentioned, between the present day (approximately 2005) and the beginning of the industrial era (approximately 1750), and is given in units of  $\text{W m}^{-2}$ .

<sup>2</sup> 90% confidence ranges are given in square brackets. Where the 90% confidence range is asymmetric about a best estimate, it is given in the form  $A [-X, +Y]$  where the lower limit of the range is  $(A-X)$  and the upper limit is  $(A+Y)$ .

<sup>3</sup> The use of “*extremely likely*” is an example of the calibrated language used in this document, it represents a 95% confidence level or higher; “*likely*” (66%) is another example. (See box TS-1).

<sup>4</sup> Estimates of RF are accompanied by both an uncertainty range (value uncertainty) and a level of scientific understanding (structural uncertainty). The value uncertainties represent the 5%-95% (90%) confidence range, 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. Climate change agents with a high level of scientific understanding are expected to have an RF that falls within their respective uncertainty ranges (see Section 2.9.1 and the TS-1 box on uncertainty for more information).

- 1 • Nitrous oxide (N<sub>2</sub>O) continues to rise approximately linearly (0.26% per year) and reached a  
2 concentration of 319 ppb in 2005, contributing an RF of +0.16 [±0.02] W m<sup>-2</sup>. Recent studies  
3 reinforce the large role of emissions from tropical regions in influencing the observed spatial  
4 concentration gradients.  
5
- 6 • Concentrations of many of the fluorine-containing Kyoto Protocol gases (hydrofluorocarbons,  
7 perfluorocarbons, SF<sub>6</sub>) have increased by large factors (between 4.3 and 1.3) between 1998 and 2005.  
8 Their total RF in 2005 was +0.017 [±0.002] W m<sup>-2</sup> and this is rapidly increasing by roughly 10% per  
9 year.  
10
- 11 • The reactive gas, OH, is a key chemical species that influences the lifetimes and thus RF values of CH<sub>4</sub>,  
12 HFCs, HCFCs and ozone; it also plays an important role in the formation of sulphate, nitrate and some  
13 organic aerosol species. Estimates of the global average OH concentration have shown no detectable net  
14 change between 1979 and 2004.  
15

16 Based on newer and better chemical transport models than in the TAR, the RF from increases in tropospheric  
17 ozone is estimated to be +0.35 [-0.1, +0.3]<sup>2</sup> W m<sup>-2</sup>, with a *medium* level of scientific understanding. There  
18 are indications of significant upward trends at low latitudes.  
19

20 The trend of greater and greater depletion of global stratospheric ozone observed during the 1980s and 1990s  
21 is no longer occurring; however, it is not yet clear whether these recent changes are indicative of ozone  
22 recovery. The RF is largely due to the destruction of stratospheric ozone by the Montreal Protocol gases and  
23 it is re-evaluated to be -0.05 [±0.10] W m<sup>-2</sup>, with a *medium* level of scientific understanding.  
24

25 Based on chemical transport model studies, the RF from increase in stratospheric water vapour due to  
26 oxidation of methane is estimated to be +0.07 [± 0.05] W m<sup>-2</sup>, with a *low* level of scientific understanding.  
27 Other potential human causes of water vapour increase that could contribute an RF are poorly understood.  
28

29 The total direct aerosol RF as derived from models and observations is estimated to be -0.5 [±0.4] W m<sup>-2</sup>,  
30 with a *medium-low* level of scientific understanding. The RF due to the cloud albedo effect (also referred to  
31 as first indirect or Twomey effect), in the context of liquid water clouds, is estimated to be -0.7 [-1.1, +0.4]<sup>2</sup>  
32 W m<sup>-2</sup>, with a *low* level of scientific understanding.  
33

- 34 • Atmospheric models have improved and many now represent all aerosol components of significance.  
35 Improved in situ, satellite and surface-based measurements have enabled verification of global aerosol  
36 models. The best estimate and uncertainty range of the total direct aerosol RF are based on a  
37 combination of modelling studies and observations.  
38
- 39 • The direct RF of the individual aerosol species is less certain than the total direct aerosol RF. The  
40 estimates are: sulphate -0.4 [±0.2] W m<sup>-2</sup>, fossil-fuel organic carbon -0.05 [±0.05] W m<sup>-2</sup>, fossil-fuel  
41 black carbon +0.2 [±0.15] W m<sup>-2</sup>, biomass burning +0.05 [±0.13] W m<sup>-2</sup>, nitrate -0.1 [±0.1] W m<sup>-2</sup>,  
42 mineral dust -0.1 [±0.2] W m<sup>-2</sup>. For biomass burning, the estimate is strongly influenced by aerosol  
43 overlying clouds. For the first time best estimates are given for nitrate and mineral dust aerosols.  
44
- 45 • Incorporation of more aerosol species and improved treatment of aerosol-cloud interactions allow a best  
46 estimate of the cloud-albedo effect. However, the uncertainty remains large. Model studies including  
47 more aerosol species or constrained by satellite observations tend to yield a relatively weaker RF. Other  
48 aspects of aerosol-cloud interactions (e.g., cloud lifetime, semi-direct effect) are not considered to be an  
49 RF (see Chapter 7).  
50

51 Land-cover changes, largely due to net deforestation, have increased the surface albedo giving an RF of -0.2  
52 [±0.2] W m<sup>-2</sup>, with a *medium-low* level of scientific understanding. Black carbon aerosol deposited on snow  
53 has reduced the surface albedo producing an associated RF of +0.1 [±0.1] W m<sup>-2</sup>, with a *low* level of  
54 scientific understanding. Other surface property changes can affect climate through processes which cannot  
55 be quantified by RF; these have a *very low* level of scientific understanding.  
56

1 Persistent linear contrails from aviation contribute an RF of  $0.01[-0.007, +0.02]^2 \text{ W m}^{-2}$ , with a *low* level of  
2 scientific understanding; the best estimate is smaller than in the TAR. No best estimates are available for the  
3 net forcing from spreading contrails and their effects on cirrus cloudiness.  
4

5 The direct RF due to increases in solar irradiance since 1750 is estimated to be  $+0.12 [-0.06, +0.18]^2 \text{ W m}^{-2}$ ,  
6 with a *low* level of scientific understanding. This RF is less than half of the TAR estimate.  
7

- 8 • The smaller RF is due to a re-evaluation of the long-term change in solar irradiance, namely a  
9 smaller increase from the Maunder Minimum to the present. However, uncertainties in the RF  
10 remain large. The total solar irradiance, monitored from space for the last three decades, reveals a  
11 well-established cycle of 0.08% (cycle min to max) with no significant trend at cycle minima.  
12
- 13 • Changes (order of a few percent) in globally averaged column ozone forced by the solar UV  
14 irradiance 11-year cycle are now better understood, but ozone profile changes are less certain.  
15 Empirical associations between solar-modulated cosmic ray ionization of the atmosphere and  
16 globally-averaged low-level cloud cover remain ambiguous.  
17

18 The global stratospheric aerosol concentrations in 2005 were at their lowest values since satellite  
19 measurements began in about 1980. This can be attributed to the absence of significant explosive volcanic  
20 eruptions since Mt. Pinatubo in 1991. Aerosols from such episodic volcanic events exert a transitory  
21 negative RF; there is, however, limited knowledge of the RF associated with eruptions prior to Mt. Pinatubo.  
22

23 The spatial patterns of RFs for non-LLGHGs (ozone, aerosol direct and cloud albedo effects, and land-use  
24 changes) have considerable uncertainties, in contrast to the relatively high confidence in that of the  
25 LLGHGs. The southern hemisphere net positive RF *very likely* exceeds that in northern hemisphere because  
26 of smaller aerosol contributions in the southern hemisphere. The RF spatial pattern is not indicative of the  
27 pattern of climate response.  
28

29 The total global-mean *surface forcing*<sup>5</sup> is *very likely* negative. By reducing the shortwave radiative flux at the  
30 surface, increases of stratospheric and tropospheric aerosols are principally responsible for the negative  
31 *surface forcing*. This is in contrast to LLGHG increases which are the principal contributors to the total  
32 positive anthropogenic RF.  
33

---

<sup>5</sup> Surface forcing is the instantaneous radiative flux change at the surface; it is a useful diagnostic tool for understanding changes in the heat and moisture surface budgets. However, unlike RF, it cannot be used for quantitative comparisons of the effects of different agents on the equilibrium global-mean surface temperature change

## 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 the *radiative forcing* (hereinafter RF) of the climate system. Discussion of the understanding of atmospheric composition changes is limited to 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 the TAR, in particular the 2002 WMO UNEP Scientific Assessment of Ozone Depletion (WMO, 2003) and the IPCC-TEAP Special Report on Safeguarding the Ozone Layer and the Global Climate System (IPCC/TEAP, 2005).

The chapter assesses anthropogenic greenhouse gas changes, aerosol changes and their impact on clouds, aviation induced contrails and cirrus changes, surface albedo changes and natural solar and volcanic mechanisms. The chapter reassesses the “*radiative forcing*” concept (Sections 2.2 and 2.8), presents spatial and temporal patterns of RF, and examines the radiative energy budget changes at the surface. .

For the long-lived-greenhouse-gases (CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, CFCs, HCFCs, HFCs, PFCs and SF<sub>6</sub>, 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. Compared to other RF agents, these trends are considerably better quantified; because of this, the chapter does not devote as much space to them as previous assessments (although the processes involved and the related budgets are further discussed in Chapter 7, Sections 7.3 and 7.4). Nevertheless LLGHGs 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 the 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 (Global 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<sup>-2</sup>) 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*”. RF is used to assess and compare the anthropogenic and natural drivers of climate change. The concept arose from early studies of the climate response to changes in solar insolation and CO<sub>2</sub>, 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 ( $\Delta T_s$ ):  $\Delta T_s = \lambda \text{RF}$ , where  $\lambda$  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 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 inadequately 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

1 of climate change comprised of “forcing” and “response”. This Chapter also uses the term *surface forcing* to  
2 refer to the instantaneous perturbation of the surface radiative balance by a forcing agent. *Surface forcing* has  
3 quite different properties than RF and should not be used to compare forcing agents (see Section 2.8.1).  
4 Nevertheless, it is a useful diagnostic, particularly for aerosols (see Sections 2.4 and 2.9).

5  
6 [INSERT FIGURE 2.1 HERE]

7  
8 Since the TAR a number of studies have investigated the relationship between RF and climate response,  
9 assessing the limitations of the RF concept; related to this there has been considerable debate whether some  
10 climate change drivers are better considered as a “forcing” or a “response” (Hansen et al., 2005; Jacob et al.,  
11 2005, Section 2.8). Emissions of forcing agents, such as LLGHGs, aerosols and aerosol precursors, ozone  
12 precursors and ozone depleting substances, are the more fundamental drivers of climate change and these  
13 emissions can be used in state-of-the-art climate models to interactively evolve forcing-agent fields along  
14 with their associated climate change. In such models some “response” is necessary to evaluate the RF. This  
15 “response” is most significant for aerosol-related cloud changes, where the tropospheric state needs to  
16 significantly alter in order to create a radiative perturbation to the climate system (Jacob et al., 2005).

17  
18 Over the paleoclimate timescales that are discussed in Chapter 6, long-term changes in forcing agents arise  
19 due to so-called “boundary condition” changes to the Earth’s climate system (such as changes in orbital  
20 parameters, ice sheets and continents). For the purposes of this chapter, these “boundary conditions” are  
21 assumed to be invariant and forcing-agent changes are considered to be *external* to the climate system. The  
22 natural RFs considered are solar changes and volcanoes; the other RF agents are all attributed to humans. For  
23 the LLGHGs it is appropriate to use observed concentrations and RF is typically calculated in offline  
24 radiative transfer schemes, using observed changes in concentration, i.e., humans are considered solely  
25 responsible for their increase. For the other climate change drivers, RF is often estimated using general  
26 circulation model (GCM) data employing a variety of methodologies (Ramaswamy et al., 2001; Stuber et al.,  
27 2001a; Tett et al., 2002; Shine et al., 2003; Hansen et al., 2005; and see Section 2.8.3). Often, alternative RF  
28 calculation methodologies that do not directly follow the TAR definition of a stratospheric-adjusted RF are  
29 used; the most important ones are illustrated in Figure 2.2. For most aerosol constituents (see Section 2.4)  
30 stratospheric adjustment has little effect on the RF, and the instantaneous RF at either the top of the  
31 atmosphere or tropopause can be substituted. For the climate change drivers discussed in Chapter 7 (Section  
32 7.5) and Section 2.5, that are not initially radiative in nature, an RF-like quantity can be evaluated allowing  
33 the tropospheric state to change: this is the zero-surface-temperature-change RF in Figure 2.2 (see Shine et  
34 al., 2003, Hansen et al., 2005 and Section 2.8.3). Other water vapour and cloud changes are considered  
35 climate feedbacks and evaluated in Chapter 8 (Section 8.6).

36  
37 [INSERT FIGURE 2.2 HERE]

38  
39 Climate change agents which necessitate changes in the tropospheric state (temperature and/or water vapour  
40 amounts) prior to causing a radiative perturbation are aerosol-cloud lifetime effects, aerosol semi-direct  
41 effects and some surface-change effects. They need to be accounted for when evaluating the overall effect of  
42 humans on climate and their radiative effects as discussed in Chapter 7 (Sections 7.2). However, in both this  
43 chapter and the assessment report they are not considered to be RFs, although the RF definition could be  
44 altered to accommodate them. Reasons for this are twofold and concern the need to be simple and pragmatic.  
45 Firstly, many GCMs have some representation of these effects inherent in their climate response and  
46 evaluation of variation in climate sensitivity between mechanisms already accounts for them (see “Efficacy”,  
47 Section 2.8.5). Secondly, the evaluation of these tropospheric state changes rely on some of the most  
48 uncertain aspects of a climate model’s response, e.g., hydrologic cycle; their radiative effects are very  
49 climate model dependent and such a dependence on climate models is what the RF concept was designed to  
50 avoid. In practice these effects can also be excluded on practical grounds – they are simply too uncertain to  
51 be adequately quantified (see Chapter 7, Section 7.5 and Sections 2.4.5 and 2.5.6).

52  
53 The RF relationship to transient climate change is not straightforward. To evaluate the overall climate  
54 response associated with a forcing agent its time evolution and its spatial and vertical structure need to be  
55 taken into account. Further, RF alone cannot be used to assess the potential climate change associated with  
56 emissions, as it does not take into account the different atmospheric lifetimes of the forcing agents. Global

1 Warming Potentials (GWPs) are one way to assess these emissions. They compare the integrated RF over a  
2 specified period (e.g., 100 years) from a unit mass pulse emission relative to CO<sub>2</sub> (see Section 2.10).

### 3 4 **2.3 Chemically and Radiatively Important Gases**

#### 5 6 **2.3.1 Atmospheric Carbon Dioxide (CO<sub>2</sub>)**

7  
8 This section discusses the instrumental measurements of CO<sub>2</sub>, documenting recent changes in atmospheric  
9 mixing ratios needed for the RF calculations presented later in the section. In addition it provides data for the  
10 preindustrial levels of CO<sub>2</sub> required as the accepted reference level for the RF calculations. For dates before  
11 about 1950 indirect measurements are relied upon. For these periods levels of atmospheric CO<sub>2</sub> are usually  
12 determined from analyses of air bubbles trapped in polar ice cores. These time periods are primarily  
13 considered in Chapter 6 of this report.

14  
15 A wide range of direct and indirect measurements confirm that its atmospheric mixing ratio has increased  
16 globally by about 100 ppm (36%) over the last 250 years, from a range of 275–285 ppm in the preindustrial  
17 era (1000–1750 AD) to 379 ppm in 2005 (see FAQ 2.1, Figure 1). During this period the absolute growth  
18 rate of CO<sub>2</sub> in the atmosphere increased substantially: the first 50 ppm increase above the preindustrial value  
19 was reached in the 1970s after more than 200 years, whereas the second 50 ppm was achieved in about 30  
20 years. In the 10 years from 1995 to 2005 atmospheric CO<sub>2</sub> increased by about 19 ppm; the highest average  
21 growth rate recorded for any decade since direct atmospheric CO<sub>2</sub> measurements began in the 1950s.

22  
23 High-precision measurements of atmospheric CO<sub>2</sub> are essential to the understanding of the carbon cycle  
24 budgets discussed in Chapter 7 (Section 7.3). The first in situ continuous measurements of atmospheric CO<sub>2</sub>  
25 made by a high-precision non-dispersive infrared gas analyser were implemented by C.D. Keeling from  
26 Scripps Institution of Oceanography (SIO) (see Chapter 1, Section 1.3). These began in 1958 at Mauna Loa,  
27 Hawaii, located at 19°N (Keeling et al., 1995). The data documented for the first time that not only was CO<sub>2</sub>  
28 increasing in the atmosphere, but also that it was modulated by cycles caused by seasonal changes in  
29 photosynthesis in the terrestrial biosphere. These measurements were followed by continuous in-situ analysis  
30 programmes at other sites in both hemispheres (Conway et al., 1994; Nakazawa et al., 1997; Langenfelds et  
31 al., 2002). In Figure 2.3 atmospheric CO<sub>2</sub> mixing ratio data at Mauna Loa in the northern hemisphere are  
32 shown with contemporaneous measurements at Baring Head, New Zealand in the southern hemisphere  
33 (Manning et al., 1997; Keeling and Whorf, 2005). These two stations provide the longest continuous records  
34 of atmospheric CO<sub>2</sub> in the northern and southern hemispheres, respectively. Remote sites such as Mauna  
35 Loa, Baring Head, Cape Grim (Tasmania) and the South Pole were chosen because air sampled at such  
36 locations shows little short term variation caused by local sources and sinks of CO<sub>2</sub> and provided the first  
37 data from which the global increase of atmospheric CO<sub>2</sub> was documented. Because CO<sub>2</sub> is a LLGHG and  
38 well mixed in the atmosphere, measurements made at such sites provide an integrated picture of large parts  
39 of the Earth including continents and city point sources. Note that this comment also applies to the other  
40 LLGHGs reported in Section 2.3.

41  
42 [INSERT FIGURE 2.3 HERE]

43  
44 In the 1980s and 1990s, it was recognized that greater coverage of CO<sub>2</sub> measurements over continental areas  
45 was required to provide the basis for estimating sources and sinks of atmospheric CO<sub>2</sub> over land as well as  
46 ocean regions. Because continuous CO<sub>2</sub> analysers are relatively expensive to maintain and require  
47 meticulous on site calibration these records are now widely supplemented by air sample flask programmes,  
48 where air is collected in glass and metal containers at a large number of continental and marine sites. After  
49 collection the filled flasks are sent to central well-calibrated laboratories for analysis. The most extensive  
50 network of international air sampling sites is operated by the National Oceanic and Atmospheric  
51 Administration's Global Monitoring Division (NOAA/GMD in the USA; formerly NOAA/CMDL). This  
52 organization collates measurements of atmospheric CO<sub>2</sub> from six continuous analyser locations as well as  
53 weekly flask air samples from a global network of almost 50 surface sites. Many international laboratories  
54 make atmospheric CO<sub>2</sub> observations and world wide databases of their measurements are maintained by the

1 Carbon Dioxide Information Analysis Center and by the World Data Centre for Greenhouse Gases in the  
2 WMO Global Atmospheric Watch programme<sup>6</sup>.

3  
4 The increases in global atmospheric CO<sub>2</sub> since the industrial revolution are mainly due to CO<sub>2</sub> emissions  
5 from the combustion of fossil fuels, gas flaring and cement production. Other sources include emissions due  
6 to land-use changes such as deforestation (Houghton, 2003) and biomass burning (Andreae and Merlet,  
7 2001; van der Werf, 2004). After entering the atmosphere, CO<sub>2</sub> exchanges rapidly with the short-lived  
8 components of the terrestrial biosphere and surface ocean, and is then redistributed on time scales of  
9 hundreds of years among all active carbon reservoirs including the long-lived terrestrial biosphere and deep  
10 ocean. The processes governing the movement of carbon between the active carbon reservoirs, climate  
11 carbon cycle feedbacks and their importance in determining the levels of CO<sub>2</sub> remaining in the atmosphere,  
12 are presented in Chapter 7 (Section 7.3) where carbon cycle budgets are discussed.

13  
14 Increase in CO<sub>2</sub> mixing ratios continues to yield the largest sustained RF of any forcing agent. The RF of CO<sub>2</sub>  
15 is a function of the change in CO<sub>2</sub> in the atmosphere over the time period under consideration. Hence a key  
16 question is “How is the CO<sub>2</sub> released from fossil fuel combustion, cement production and land cover change  
17 distributed amongst the atmosphere, oceans and terrestrial biosphere?” This partitioning has been  
18 investigated using a variety of techniques. Amongst the most powerful of these are measurements of the  
19 carbon isotopes in CO<sub>2</sub> as well as high precision measurements of atmospheric O<sub>2</sub> content. The carbon  
20 contained in CO<sub>2</sub> has two naturally occurring stable isotopes denoted <sup>12</sup>C and <sup>13</sup>C. The first of these, <sup>12</sup>C, is  
21 the most abundant isotope at about 99% followed by <sup>13</sup>C at about 1%. CO<sub>2</sub> emitted from coal, gas and oil  
22 combustion and land clearing has <sup>13</sup>C/<sup>12</sup>C isotopic ratios that are less than those in atmospheric CO<sub>2</sub> and each  
23 carries a signature related to its source. Thus, as shown in Prentice et al. (2001), when CO<sub>2</sub> from fossil fuel  
24 combustion enters the atmosphere, the <sup>13</sup>C/<sup>12</sup>C isotopic ratio in atmospheric CO<sub>2</sub> decreases at a predictable  
25 rate consistent with emissions of CO<sub>2</sub> from fossil origin. Note that changes in the <sup>13</sup>C/<sup>12</sup>C ratio of  
26 atmospheric CO<sub>2</sub> are also caused by other sources and sinks but the changing isotopic signal due to CO<sub>2</sub>  
27 from fossil fuel combustion can be resolved from the other components (Francey et al., 1995). These  
28 changes can easily be measured using modern isotope ratio mass spectrometry which has the capability of  
29 measuring <sup>13</sup>C/<sup>12</sup>C in atmospheric CO<sub>2</sub> to better than 1 part in 10<sup>5</sup> (Ferretti et al., 2000). Data presented in  
30 Figure 2.3 for the <sup>13</sup>C/<sup>12</sup>C ratio of atmospheric CO<sub>2</sub> at Mauna Loa show a decreasing ratio, contrary to trends  
31 in both fossil fuel CO<sub>2</sub> emissions and atmospheric CO<sub>2</sub> mixing ratios (Andres et al., 2000; Keeling et al.,  
32 2005).

33  
34 Atmospheric oxygen measurements provide a powerful and independent method of determining the  
35 partitioning of CO<sub>2</sub> between the oceans and land (Keeling et al., 1996). Atmospheric O<sub>2</sub> and CO<sub>2</sub> changes are  
36 inversely coupled during plant respiration and photosynthesis. Also, during the process of combustion O<sub>2</sub> is  
37 removed from the atmosphere producing a signal that decreases as atmospheric CO<sub>2</sub> increases on a molar  
38 basis (Figure 2.3). Measuring changes in atmospheric O<sub>2</sub> is technically challenging because of the difficulty  
39 of resolving changes at the ppm level in a background mixing ratio of roughly 209,000 ppm. These  
40 difficulties were first overcome by Keeling and Shertz (1992) who used an interferometric technique to  
41 show that it is possible to track both seasonal cycles and the ppm-level decline of oxygen in the atmosphere  
42 (Figure 2.3). Recent work by Manning and Keeling (2006) indicates that atmospheric O<sub>2</sub> is decreasing at a  
43 faster rate than CO<sub>2</sub> is increasing which demonstrates the importance of the oceanic carbon sink.  
44 Measurements of both the <sup>13</sup>C/<sup>12</sup>C ratio in atmospheric CO<sub>2</sub> and atmospheric O<sub>2</sub> levels are valuable tools used  
45 to determine the distribution of fossil fuel derived CO<sub>2</sub> amongst the active carbon reservoirs, as discussed in  
46 Chapter 7 (Section 7.3). In Figure 2.3, recent measurements in both hemispheres are shown to emphasize the  
47 strong linkages between atmospheric CO<sub>2</sub> increases, O<sub>2</sub> decreases, fossil fuel consumption and the <sup>13</sup>C/<sup>12</sup>C  
48 ratio of atmospheric CO<sub>2</sub>.

49  
50 From 1990 to 1999, a period reported in Prentice et al. (2001), the emission rate due to fossil fuel burning  
51 and cement production increased irregularly from 6.1 to 6.5 GtC yr<sup>-1</sup> or about 0.7% yr<sup>-1</sup>. From 1999 to 2005  
52 however, the emission rate rose systematically from 6.5 to 7.8 GtC yr<sup>-1</sup> (BP World Energy, 2006; Marland et  
53 al., 2006) or about 3.0% yr<sup>-1</sup> representing a period of higher emissions and growth in emissions than those  
54 considered in the TAR (see Figure 2.3). CO<sub>2</sub> emissions due to global annual fossil fuel combustion and  
55 cement manufacture combined have increased by 70% over the last 30 years (Marland et al., 2006). The

<sup>6</sup> CDIAC, <http://cdiac.esd.ornl.gov/>; WDCGG, <http://gaw.kishou.go.jp/wdcgg.html>

1 relationship between increases in atmospheric CO<sub>2</sub> mixing ratios and emissions have been tracked using a  
2 scaling factor known as the apparent “airborne fraction” defined as the ratio of the annual increase in  
3 atmospheric CO<sub>2</sub> to the CO<sub>2</sub> emissions from annual fossil fuel and cement manufacture combined (Keeling et  
4 al., 1995). On decadal scales this fraction has averaged about 60% since the 1950s. Assuming emissions of 7  
5 GtC yr<sup>-1</sup> and an airborne fraction remaining at about 60%, Hansen and Sato (2004) predicted that the  
6 underlying long term global atmospheric CO<sub>2</sub> growth rate will be about 1.9 ppm yr<sup>-1</sup> a value consistent with  
7 observations over the 1995-2005 decade.

8  
9 Carbon dioxide emissions due to land use changes during the 1990s are estimated as 0.5–2.7 GtC yr<sup>-1</sup>  
10 (Chapter 7, Section 7.3, Table 7.2), contributing 6%-39% of the CO<sub>2</sub> growth rate (Brovkin et al., 2004).  
11 Prentice et al. (2001) cited an inventory-based estimate that land use change resulted in net emissions of 121  
12 GtC between 1850 and 1990, after Houghton (1999; 2000). The estimate for this period was revised upwards  
13 to 134 GtC by Houghton (2003), mostly due to an increase in estimated emissions prior to 1960. Houghton  
14 (2003) also extended the inventory emissions estimate to 2000 giving cumulative emissions of 156 GtC since  
15 1850. In carbon cycle simulations by Brovkin et al. (2004) and Matthews et al. (2004), land use change  
16 emissions contributed 12–35 ppm of the total CO<sub>2</sub> rise from 1850 to 2000 (Table 2.8). Historical changes in  
17 land cover are discussed in Section 2.5.2, and the CO<sub>2</sub> budget over the 1980s and 1990s is discussed further  
18 in Chapter 7 (Section 7.3).

19  
20 In 2005, the global mean average CO<sub>2</sub> mixing ratio for the SIO network of 9 sites was 378.75 ± 0.13 ppm  
21 and for the NOAA/GMD network of 40 sites was 378.76 ± 0.05 ppm yielding a global average of almost 379  
22 ppm. For both networks only sites in the remote marine boundary layer are used and high altitude locations  
23 are not included. The Mauna Loa site for example is excluded due to an “altitude effect” of about 0.5 ppm.  
24 Also the 2005 values are still pending final reference gas calibrations used to measure the samples.

25  
26 New measurements of CO<sub>2</sub> from Antarctic ice and firn (MacFarling Meure et al., 2006) update and extend  
27 those from Etheridge et al. (1996) to 0 AD. The CO<sub>2</sub> mixing ratio in 1750 was 277 ± 1.2 ppm<sup>7</sup>. This record  
28 shows variations between 272 and 284 ppm before 1800 and that CO<sub>2</sub> mixing ratios dropped by 5–10 ppm  
29 between 1600 and 1800 (see Chapter 6, Section 6.3). The RF calculations usually take 1750 as the  
30 preindustrial index (e.g., the TAR and this report). Therefore, using 1750 may slightly over-estimate the RF,  
31 as the changes in the mixing ratios of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O after the end of this naturally cooler period may not  
32 be solely attributable to anthropogenic emissions. Using 1860 as an alternative start date for the RF  
33 calculations would reduce the LLGHG RF by roughly 10%. For the RF calculation the data from Law Dome  
34 ice cap in the Antarctic are used because they show the highest age resolution (approximately 10 years) of  
35 any ice core record in existence. In addition the high precision data from the cores are connected to direct  
36 observational records of atmospheric CO<sub>2</sub> from Cape Grim, Tasmania.

37  
38 The simple formulae for RF of the LLGHG quoted in Ramaswamy et al. (2001) are still valid. These  
39 formulae are based on global RF calculations where clouds, stratospheric adjustment and solar absorption  
40 were included and give an RF of 3.7 W m<sup>-2</sup> for a doubling in the CO<sub>2</sub> mixing ratio. (The formula used for the  
41 CO<sub>2</sub> RF calculation in this chapter is the IPCC (1990) expression as revised in the TAR. Note that for CO<sub>2</sub>  
42 RF increases logarithmically with mixing ratio.) Collins et al. (2006) performed a comparison of 5 detailed  
43 line-by-line models and 20 GCM radiation schemes. The spread of line-by-line model results were consistent  
44 with the ±10% uncertainty estimate for the LLGHG RFs adopted in Ramaswamy et al. (2001) and a similar  
45 ±10% for the 90% confidence interval is adopted here. However, it is also important to note that these  
46 relatively small uncertainties are not always achievable when incorporating the LLGHG forcings into GCMs.  
47 For example, both Collins et al. (2006) and Forster and Taylor (2006) found that GCM radiation schemes  
48 could have inaccuracies of around 20% in their total LLGHG RF (see also Section 2.3.2 and Chapter 10,  
49 Section 10.2).

50  
51 Using the global average value of 379 ppm for atmospheric CO<sub>2</sub> in 2005 gives an RF of 1.66 ± 0.17 W m<sup>-2</sup>; a  
52 contribution that dominates that of all other forcing agents considered in this chapter. This is an increase of  
53 13–14% over the value reported for 1998 in Ramaswamy et al. (2001). This change is solely due to increases  
54 in atmospheric CO<sub>2</sub> and is also much larger than the RF changes due to other agents. In the decade 1995 to

---

<sup>7</sup> For consistency with the TAR the preindustrial value of 278 ppm is retained in the CO<sub>2</sub> RF calculation.

1 2005, the RF due to CO<sub>2</sub> increased by about 0.28 W m<sup>-2</sup> (20%), an increase greater than that calculated for  
2 any decade since at least 1800 (see Chapter 6 6, Section 6.6; also see FAQ 2.1, Figure 1).

3  
4 Table 2.1, summarizes the present day mixing ratios and RF for the LLGHGs, and indicates changes since  
5 1998. The RF from CO<sub>2</sub> and that from the other LLGHGs have a *high* level of scientific understanding  
6 (Table 2.11). Note that the uncertainty in RF is almost entirely due to radiative transfer assumptions and not  
7 mixing ratio estimates, therefore trends in RF can be more accurately determined than the absolute RF. From  
8 Table 2.8, the contribution from land-use change to the present CO<sub>2</sub> RF is *likely* to be about 0.4 Wm<sup>-2</sup> (since  
9 1850). This implies that fossil fuel and cement production have *likely* contributed about three-quarters of the  
10 current RF.

11  
12 [INSERT TABLE 2.1 HERE]

### 13 14 2.3.2 Atmospheric Methane (CH<sub>4</sub>)

15  
16 This section describes the current global measurement programmes for atmospheric methane (CH<sub>4</sub>), which  
17 provide the data required for the understanding of its budget and for the calculation of its RF. In addition it  
18 provides data for the preindustrial levels of CH<sub>4</sub> required as the accepted reference level for these  
19 calculations. Detailed analyses of methane budgets and its biogeochemistry are presented in Chapter 7  
20 (Section 7.4).

21  
22 Methane has the second largest RF of the LLGHGs after CO<sub>2</sub> (Ramaswamy et al., 2001). Over the last  
23 650,000 years ice core records indicate that the abundance of methane in the earth's atmosphere has varied  
24 from lows of about 400 ppb during glacial periods to highs of about 700 ppb during inter-glacials (Spahni et  
25 al., 2005) with a single measurement from the Vostok core reaching about 770 ppb (see Chapter 6, Figure  
26 6.3).

27  
28 In 2005 the global average abundance of CH<sub>4</sub> measured at the network of 40 surface air flask sampling sites  
29 operated by NOAA/GMD in both hemispheres was 1774.62 ± 1.22<sup>8</sup> ppb. This network of sites is the most  
30 geographically extensive operated by any laboratory and it is important to note that the calibration scale used  
31 by it has changed since the TAR (Dlugokencky et al., 2005). The new scale (known as NOAA04) increases  
32 all previously reported CH<sub>4</sub> mixing ratios from NOAA/GMD by about 1% bringing them into much closer  
33 agreement with the AGAGE network. This scale will be used by laboratories participating in the WMO's  
34 Global Atmosphere Watch Programme as a "common reference". Atmospheric CH<sub>4</sub> is also monitored at 5  
35 sites in the northern and southern hemispheres by the AGAGE network. This group uses automated systems  
36 to make 36 methane measurements a day at each site and the mean for 2005 was 1774.03 ± 1.68<sup>9</sup> ppb with  
37 calibration and methods described by Cunnold et al. (2002). For the NOAA/GMD network, the 90%  
38 confidence interval is calculated with a Monte Carlo technique, which only accounts for the uncertainty due  
39 to the distribution of sampling sites. For both networks, only sites in the remote marine boundary layer are  
40 used and continental sites are not included. Global databases of atmospheric CH<sub>4</sub> measurements for these  
41 and other CH<sub>4</sub> measurement programmes (e.g., Japanese, European and Australian) are maintained by the  
42 Carbon Dioxide Information Analysis Center (<http://cdiac.esd.ornl.gov/>) and by the World Data Centre for  
43 Greenhouse Gases in the Global Atmospheric Watch programme (<http://gaw.kishou.go.jp/wdcgg.html>).

44  
45 Present atmospheric levels of CH<sub>4</sub> are unprecedented in at least the last 650,000 years (Spahni et al., 2005).  
46 Direct atmospheric measurements of the gas made at a wide variety of sites in both hemispheres over the last  
47 25 years show that, although the abundance of CH<sub>4</sub> has increased by about 30% during that time, its growth  
48 rate has decreased substantially from highs of greater than 1% yr<sup>-1</sup> in the late 1970s and early 1980s (Blake  
49 and Rowland, 1988) to lows of close to zero towards the end of the 1990s (Dlugokencky et al., 1998;  
50 Simpson et al., 2002). The slowdown in the growth rate began in the 1980s, decreasing from 14 ppb yr<sup>-1</sup>  
51 (about 1% yr<sup>-1</sup>) in 1984 to close to zero during 1999–2005, for the network of surface sites maintained by  
52 NOAA/GMD (Dlugokencky et al., 2003). Measurements by Lowe et al. (2004) for sites in the southern

---

<sup>8</sup> The 90% confidence range quoted is from the normal 1σ errors for trace gas measurements assuming a normal distribution, i.e. multiplying by a factor of 1.645.

<sup>9</sup> The 90% confidence range quoted is from the normal 1σ errors for trace gas measurements assuming a normal distribution, i.e. multiplying by a factor of 1.645.

1 hemisphere and Cunnold et al. (2002) for the network of GAGE/AGAGE sites show similar features. A key  
2 feature in the global growth rate of CH<sub>4</sub> is its current interannual variability with growth rates ranging from a  
3 high of 14 ppb yr<sup>-1</sup> in 1998 to less than zero in 2001, 2004 and 2005. (Figure 2.4)

4  
5 [INSERT FIGURE 2.4 HERE]

6  
7 The reasons for the decrease in the atmospheric CH<sub>4</sub> growth rate and the implications for future changes in  
8 its atmospheric burden are not understood (Prather et al., 2001) but are clearly related to changes in the  
9 imbalance between CH<sub>4</sub> sources and sinks. Most methane is removed from the atmosphere by reaction with  
10 the hydroxyl free radical (OH) which is produced photochemically in the atmosphere. The role of OH in  
11 controlling atmospheric CH<sub>4</sub> levels is discussed in Section 2.3.5. Other minor sinks include reaction with  
12 free chlorine (Platt et al., 2004; Allan et al., 2005), destruction in the stratosphere and soil sinks (Born et al.,  
13 1990).

14  
15 The total global CH<sub>4</sub> source is relatively well known but the strength of each source component and their  
16 trends are not. As detailed in Chapter 7 (Section 7.4) the sources are mostly biogenic and include wetlands,  
17 rice agriculture, biomass burning, and ruminant animals. Methane is also emitted by various industrial  
18 sources including fossil fuel mining and distribution. Prather et al. (2001) documented a large range in  
19 “bottom up” estimates for the global source in CH<sub>4</sub>. New source estimates published since then are  
20 documented in Chapter 7 (Table 7.6). However, as reported by Bergamaschi et al. (2005) national  
21 inventories based on “bottom up” studies can grossly underestimate emissions and “top down”  
22 measurement-based assessments of reported emissions will be required for verification. Keppler et al., 2006  
23 reported the discovery of emissions of CH<sub>4</sub> from living vegetation and estimated that this contributed 10–  
24 30% of the global CH<sub>4</sub> source. This work extrapolates limited measurements to a global source and has not  
25 yet been confirmed by other laboratories, but lends some support to spaceborne observations of CH<sub>4</sub> plumes  
26 above tropical rainforests reported by Frankenberg et al. (2005). That such a potentially large source of CH<sub>4</sub>  
27 could have been missed highlights the large uncertainties involved in current “bottom up” estimates of  
28 components of the global source (see Chapter 7, Section 7.4).

29  
30 Several wide ranging hypothesis have been put forward to explain the reduction in the growth rate of CH<sub>4</sub>  
31 and its variability. For example, Hansen et al. (2000) considered that economic incentives have led to a  
32 reduction in anthropogenic CH<sub>4</sub> emissions. The negligible long-term change in its main sink (OH) (see  
33 Section 2.3.5 and Figure 2.8) implies that CH<sub>4</sub> emissions are not increasing. Similarly, Dlugokencky et al.  
34 (1998) and Francey et al. (1999) suggest that the slowdown in the growth rate reflects a stabilization of CH<sub>4</sub>  
35 emissions, given that the observations are consistent with stable emissions and lifetime since 1982.

36  
37 Relatively large anomalies occurred in the growth rate during 1991 and 1998, with peak values reaching 15  
38 and 14 ppb yr<sup>-1</sup> respectively (about 1% yr<sup>-1</sup>). The anomaly in 1991 was followed by a dramatic drop in the  
39 growth rate in 1992 and has been linked with the Pinatubo volcanic eruption in June 1991 which injected  
40 large amounts of ash and SO<sub>2</sub> into the lower stratosphere of the tropics with subsequent impacts on tropical  
41 photochemistry and the removal of CH<sub>4</sub> by atmospheric OH (Bekki et al., 1994; Dlugokencky et al., 1996).  
42 Lelieveld et al. (1998) and Walter et al. (2001a; 2001b) proposed that lower temperatures and lower  
43 precipitation in the aftermath of the Pinatubo eruption could have suppressed CH<sub>4</sub> emissions from wetlands.  
44 At this time, and in parallel with the growth rate anomaly in the methane mixing ratio an anomaly was  
45 observed in methane's <sup>13</sup>C/<sup>12</sup>C ratio at surface sites in the southern hemisphere. This was attributed to a  
46 decrease in emissions from an isotopically heavy source such as biomass burning (Lowe et al., 1997; Mak et  
47 al., 2000), although these data were not confirmed by lower frequency measurements from the same period  
48 made by Francey et al. (1999).

49  
50 For the relatively large increase in the CH<sub>4</sub> growth rate reported for 1998, Dlugokencky et al. (2001) suggest  
51 that wetland and boreal biomass burning sources may have contributed to the anomaly, noting that 1998 was  
52 the warmest year globally since surface instrumental temperature records began. Using an inverse method,  
53 Chen and Prinn (2006) attribute the same event primarily to increased wetland and rice region emissions and  
54 secondarily to biomass burning. The same conclusion is reached by Morimoto et al. (2006) who used carbon  
55 isotopic measurements of CH<sub>4</sub> to constrain the relative contributions of biomass burning (1/3) and wetlands  
56 (2/3) to the increase.

1 On the basis of ice core measurements of methane (Etheridge et al., 1998) the preindustrial global value for  
2 CH<sub>4</sub> from 1700 to 1800 was 715 ± 4 ppb (it was also 715 ± 4 ppb in 1750) thus providing the reference level  
3 for the RF calculation. This takes into account the inter-polar difference in CH<sub>4</sub> as measured from Greenland  
4 and Antarctic ice cores.

5  
6 The RF due to changes in CH<sub>4</sub> mixing ratio is calculated with the simplified yet still valid expression for CH<sub>4</sub>  
7 given in Ramaswamy et al. (2001). The change in the CH<sub>4</sub> mixing ratio from 715 ppb in 1750 to 1774 ppb  
8 (the average mixing ratio from the AGAGE and GMD networks in 2005) gives an RF of 0.48 ± 0.05 W m<sup>-2</sup>,  
9 ranking CH<sub>4</sub> as the second highest RF of the LLGHGs after CO<sub>2</sub> (Table 2.1). The uncertainty range in  
10 mixing ratios for the present day represents intra-annual variability, which is not included in the preindustrial  
11 uncertainty estimate derived solely from ice-core sampling precision. The estimate for the RF due to CH<sub>4</sub> is  
12 the same as in Ramaswamy et al. (2001) despite the small increase in its mixing ratio. The spectral  
13 absorption by methane is overlapped to some extent by N<sub>2</sub>O lines (taken into account in the simplified  
14 expression). Taking the overlapping lines into account using current N<sub>2</sub>O mixing ratios instead of  
15 preindustrial mixing ratios (as in Ramaswamy et al., 2001), reduces the current RF due to CH<sub>4</sub> by 1%.

16  
17 Collins et al. (2006) confirm that line by line-models agree extremely well for the calculation of clear-sky  
18 instantaneous RF from CH<sub>4</sub> and N<sub>2</sub>O when the same atmospheric background profile is used. However,  
19 GCM radiation schemes were found to be in poor agreement with the line-by line models and errors of over  
20 50% were possible for CH<sub>4</sub>, N<sub>2</sub>O and the CFCs. In addition a small effect from the absorption of solar  
21 radiation was found with the line-by-line models, which the GCMs did not include (Chapter 10, Section  
22 10.2).

### 23 24 2.3.3 Other Kyoto Protocol Gases (N<sub>2</sub>O, HFCs, PFCs, and SF<sub>6</sub>)

25  
26 At the time of the TAR, nitrous oxide (N<sub>2</sub>O) had the fourth largest RF among the LLGHGs behind CO<sub>2</sub>, CH<sub>4</sub>  
27 and CFC-12. The TAR quoted an atmospheric N<sub>2</sub>O abundance of 314 ppb in 1998, an increase of 44 ppb  
28 from its preindustrial level of around 270 ± 7 ppb, which gave an RF of 0.15 ± 0.02 W m<sup>-2</sup>. This RF is  
29 affected by atmospheric methane levels due to overlapping absorptions. As nitrous oxide is also the major  
30 source of ozone-depleting NO and NO<sub>2</sub> in the stratosphere it is routinely reviewed in the ozone assessments;  
31 the most recent assessment (Montzka et al., 2003) recommended a lifetime of 114 years for N<sub>2</sub>O. TAR  
32 pointed out large uncertainties in the major soil, agricultural, combustion and oceanic sources of N<sub>2</sub>O. Given  
33 these emission uncertainties, its observed rate of increase of 0.2 to 0.3% per year was not inconsistent with  
34 its better quantified major sinks (principally stratospheric destruction). The primary driver for the industrial-  
35 era increase of N<sub>2</sub>O was concluded to be enhanced microbial production in expanding and fertilized  
36 agricultural lands.

37  
38 Ice-core data for N<sub>2</sub>O have been reported extending back 2000 years and more before present (MacFarling  
39 Meure et al., 2006 and Chapter 6, Section 6.6). These data, as for CO<sub>2</sub> and CH<sub>4</sub>, show relatively little  
40 changes in mixing ratios over the first 1800 years of this record, and then exhibit a relatively rapid rise (see  
41 Figure FAQ2.1). Since 1998 atmospheric N<sub>2</sub>O levels have steadily risen to 319 ± 0.12 ppb in 2005, and  
42 levels have now been increasing approximately linearly (at around 0.26% per year) for the past few decades  
43 (Figure 2.5). A change in the N<sub>2</sub>O mixing ratio from 270 ppb in 1750 to 319 ppb in 2005 results in an RF of  
44 0.16 ± 0.02 W m<sup>-2</sup>, calculated using the simplified expression given in Ramaswamy et al. (2001). The RF  
45 has increased by 11% since the time of the TAR (Table 2.1). As CFC-12 levels slowly decline (see Section  
46 2.3.4), N<sub>2</sub>O should, with its current trend, take over third place in the LLGHG RF ranking.

47  
48 [INSERT FIGURE 2.5 HERE]

49  
50 Since the TAR, understanding of regional N<sub>2</sub>O fluxes has improved. The results of various studies which  
51 quantified the global N<sub>2</sub>O emissions from coastal upwelling areas, continental shelves, estuaries and rivers  
52 suggest that these coastal areas contribute 0.3–6.6 TgN(in N<sub>2</sub>O) yr<sup>-1</sup> or 7–61% of the total oceanic emissions  
53 (Bange et al, 1996; Nevison et al, 2004b; Kroeze et al, 2005) (see also Chapter 7, Section 7.4). Using inverse  
54 methods and AGAGE Ireland measurements, Manning et al. (2003) have estimated European Union  
55 emissions of 0.9 ± 0.1 Tg N<sub>2</sub>O-N yr<sup>-1</sup> that agree well with the United Nations FCCC inventory (0.8 ± 0.1 Tg  
56 N<sub>2</sub>O-N yr<sup>-1</sup>). Melillo et al. (2001) provided evidence from Brazilian land-use sequences that the conversion  
57 of tropical forest to pasture leads to an initial increase but a later decline in emissions of N<sub>2</sub>O relative to the

1 original forest. They also deduced that Brazilian forest soils alone contribute about 10% of total global N<sub>2</sub>O  
2 production. Estimates of N<sub>2</sub>O sources and sinks using observations and inverse methods had earlier implied  
3 that a large fraction of global N<sub>2</sub>O emissions in 1978–1988 were tropical: specifically 20–29% in 0°–30°S  
4 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).  
5 These estimates were uncertain due to their significant sensitivity to assumed troposphere-stratosphere  
6 exchange rates that strongly influence interhemispheric gradients. Hirsch et al. (2006) used inverse  
7 modelling to estimate significantly lower emissions in 30°S–90°S (0–4%) and higher emissions in 0°–30°N  
8 (50–64%) than Prinn et al. (1990) during 1998–2001, with 26–36% from the oceans. The stratosphere is also  
9 proposed to play an important role in the seasonal cycles of N<sub>2</sub>O (Nevison et al., 2004a). For example, its  
10 well-defined seasonal cycle in the southern hemisphere has been interpreted as resulting from the net effect  
11 of seasonal oceanic outgassing of microbially-produced N<sub>2</sub>O, stratospheric intrusion of low-N<sub>2</sub>O air, and  
12 other processes (Nevison et al., 2005). These authors also estimated a southern ocean (30°–90°S) source of  
13 0.9 Tg N<sub>2</sub>O-N yr<sup>-1</sup>, or about 5% of the global total. The complex seasonal cycle in the northern hemisphere is  
14 more difficult to reconcile with seasonal variations in the northern latitude soil sources and stratospheric  
15 intrusions (Prinn et al., 2000; Liao et al., 2004). The destruction of N<sub>2</sub>O in the stratosphere causes  
16 enrichment of its heavier isotopomers and isotopologues providing a potential method to differentiate  
17 stratospheric and surface flux influences on tropospheric N<sub>2</sub>O (Morgan et al., 2004).

18  
19 Human-made PFCs, HFCs, and SF<sub>6</sub> are very effective absorbers of infrared radiation, so that even small  
20 amounts of these gases contribute significantly to the RF of the climate system. The observations and global  
21 cycles of the major HFCs, PFCs and SF<sub>6</sub> have recently been reviewed in Chapter 2 of IPCC/TEAP (2005):  
22 Velders et al. (2005), and this section only provides a brief review and an update for these species. Table 2.1  
23 shows the present mixing ratio and recent trends in the halocarbons; it also shows RF. Absorption spectra of  
24 most halocarbons reviewed here and in the following section are characterized by strongly overlapping  
25 spectral lines that are not resolved at tropospheric pressures and temperatures, and there is some uncertainty  
26 in cross-section measurements. Apart from the uncertainties stemming from the cross-sections themselves,  
27 differences in the radiative flux calculations can arise from the spectral resolution used, tropopause heights,  
28 vertical, spatial and seasonal distributions of the gases, cloud cover, and how stratospheric temperature  
29 adjustments are performed. IPCC/TEAP (2005) concludes that the discrepancy in the RF calculation for  
30 different halocarbons, associated with uncertainties in the radiative transfer calculation and the cross-  
31 sections, can reach 40%. Studies reviewed in IPCC/TEAP (2005) for the more abundant HFCs show that an  
32 agreement better than 12% can be reached for these when the calculation conditions are better constrained  
33 (see Section 2.10.2).

34  
35 The HFCs of industrial importance have lifetimes in the range 1.4–270 years. The HFCs with the largest  
36 observed mole fractions in 1998 as reported in the TAR were, in order: HFC-23 (CHF<sub>3</sub>), HFC-134a  
37 (CF<sub>3</sub>CH<sub>2</sub>F) and HFC-152a (CH<sub>3</sub>CHF<sub>2</sub>). In 2005 the observed mixing ratios of the major HFCs in the  
38 atmosphere were 35 ppt for HFC-134a, 17.5 ppt for HFC-23 (2003 value), 3.7 ppt for HFC-125, and 3.9 ppt  
39 for HFC-152a (Table 2.1). Within the uncertainties in calibration and emissions estimates the observed  
40 mixing ratios of the HFCs in the atmosphere can be explained by the anthropogenic emissions.  
41 Measurements are available from GMD (Thompson et al., 2004) and AGAGE (Prinn et al., 2000; O'Doherty  
42 et al., 2004; Prinn et al., 2005b) networks as well as from UEA studies in Tasmania (updated from Oram et  
43 al., 1998; Oram, 1999). These data, summarized in Figure 2.6, show a continuation of positive HFC trends  
44 and increasing latitudinal gradients (larger trends in the northern hemisphere) due to their predominantly  
45 northern hemispheric sources. The air-conditioning refrigerant, HFC-134a, is increasing at a rapid rate in  
46 response to its growing emissions arising from its role as a replacement for some CFC refrigerants. With a  
47 lifetime of about 14 years, its current trends are determined primarily by its emissions and secondarily by its  
48 atmospheric destruction. Emissions of HFC-134a estimated from atmospheric measurements are in  
49 approximate agreement with industry estimates (Huang and Prinn, 2002; O'Doherty et al., 2004).  
50 IPCC/TEAP (2005) reported that global HFC-134a emissions started rapidly increasing in the early 1990s  
51 and that in Europe, sharp increases in emissions are noted for HFC-134a over 1995–1998 and for HFC-152a  
52 over 1996–2000, with some levelling off through 2003. The concentration of the foam-blower HFC-152a,  
53 with a lifetime of only about 1.5 years, is rising approximately exponentially, with the effects of increasing  
54 emissions being only partly offset by its rapid atmospheric destruction. HFC-23 has a very long atmospheric  
55 lifetime (approximately 270 years) and is mainly produced as a by-product of HCFC-22 (CHF<sub>2</sub>Cl)  
56 production. Its concentrations are rising approximately linearly, driven by these emissions, with its

1 destruction being only a minor factor in its budget. There are also smaller but rising concentrations of HFC-  
2 125 ( $\text{CHF}_2\text{CF}_3$ ) and HFC-143a ( $\text{CH}_3\text{CF}_3$ ), which are both refrigerants.

3  
4 [INSERT FIGURE 2.6 HERE]

5  
6 The PFCs, mainly  $\text{CF}_4$  (PFC-14) and  $\text{C}_2\text{F}_6$  (PFC-116), and  $\text{SF}_6$  have very large radiative efficiencies and  
7 lifetimes in the range 1,000–50,000 years (see Section 2.10, Table 2.14), and make an essentially permanent  
8 contribution to RF. The  $\text{SF}_6$  and  $\text{C}_2\text{F}_6$  concentrations and RF have increased by over 20% since the TAR  
9 (Table 2.1, Figure 2.6), but  $\text{CF}_4$  concentrations have not been updated since 1997. Both anthropogenic and  
10 natural sources of  $\text{CF}_4$  are important to explain its observed atmospheric abundance. These PFCs are  
11 produced as by-products of traditional aluminium production, among other activities. The  $\text{CF}_4$  concentrations  
12 have been increasing linearly since about 1960 and  $\text{CF}_4$  has a natural source that accounts for about one half  
13 of its current atmospheric content (Harnisch et al., 1996). Sulphur hexafluoride ( $\text{SF}_6$ ) is produced for use as  
14 an electrical insulating fluid in power distribution equipment and also deliberately released as an essentially  
15 inert tracer to study atmospheric and oceanic transport processes. Its concentration was 4.2 ppt in 1998  
16 (TAR) and has continued to increase linearly over the past decade implying that emissions are approximately  
17 constant. Its very long lifetime ensures that its emissions accumulate essentially unabated in the atmosphere.

### 18 19 **2.3.4 Montreal Protocol Gases (CFCs and HCFCs)**

20  
21 The Montreal Protocol for protection of the ozone layer regulates many radiatively powerful greenhouse  
22 gases for the primary purpose of lowering stratospheric chlorine and bromine concentrations. These gases  
23 include the CFCs, HCFCs, chlorocarbons, bromocarbons, and halons. Observations and global cycles of  
24 these gases have been recently reviewed in detail in Chapter 1 of the 2002 WMO-UNEP Scientific  
25 Assessment of Ozone Depletion (WMO, 2003) and IPCC/TEAP (2005). The discussion here focuses on  
26 developments since these reviews and on those gases that contribute most to RF rather than to halogen  
27 loading. Using observed 2005 concentrations, the Montreal Protocol gases have contributed 12% ( $0.320 \text{ W}$   
28  $\text{m}^{-2}$ ) to the direct RF of all LLGHGs and 95% to the halocarbon RF (Table 2.1). This contribution is  
29 dominated by the CFCs. The effect of the Montreal Protocol on these gases has been substantial.  
30 IPCC/TEAP (2005) concluded that the combined  $\text{CO}_2$ -equivalent emissions of CFCs, HCFCs, and HFCs  
31 decreased from a peak of about  $7.5 \text{ GtCO}_2\text{-eq yr}^{-1}$  in the late 1980s to about  $2.5 \text{ GtCO}_2\text{-eq yr}^{-1}$  by the year  
32 2000, corresponding to about 10% of that year's  $\text{CO}_2$  emission due to global fossil fuel burning.

33  
34 Measurements of the CFCs and HCFCs, summarized in Figure 2.6, are available from the AGAGE network  
35 (Prinn et al., 2000; Prinn et al., 2005b) and the GMD network (Montzka et al., 1999 updated; Thompson et  
36 al., 2004). Certain flask measurements are also available from UCI (Blake et al., 2001 updated) and UEA  
37 (Oram et al., 1998; Oram, 1999 updated). Two of the major CFCs (CFC-11, CFC-113) have both been  
38 decreasing in the atmosphere since the mid-1990s. While their emissions have decreased very substantially  
39 in response to the Montreal Protocol, their long lifetimes of around 45 and 85 years, respectively, mean that  
40 their sinks can reduce their levels only at about 2% and 1% per year, respectively. Nevertheless the effect of  
41 the Montreal Protocol has been to substantially reduce the growth of the halocarbon RF which increased  
42 rapidly from 1950 until about 1990. The other major CFC ( $\text{CF}_2\text{Cl}_2$  or CFC-12), which is the third most  
43 important LLGHG, is finally reaching a plateau in its atmospheric levels (emissions equal loss) and may  
44 have peaked in 2003. Its 100-year lifetime means that it can decrease only at about 1% per year even when  
45 emissions are zero. The levelling off for CFC-12 and approximately linear downward trends for CFC-11 and  
46 CFC-113 continue. Latitudinal gradients for all three are very small and decreasing as expected. The  
47 combined CFC and HCFC RF has been slowly declining since 2003. Note that the 1998 concentrations of  
48 CFC-11 and CFC-12 were overestimated in Table 6.1 of the TAR. This means that the total halocarbon RF  
49 quoted for 2005 in Table 2.1 ( $0.337 \text{ W m}^{-2}$ ) is slightly smaller than the  $0.34 \text{ W m}^{-2}$  quoted in the TAR. In  
50 fact the measurements indicate a small 1% rise in the total halocarbon RF since the time of the TAR (Table  
51 2.1).

52  
53 The major solvent, methyl chloroform ( $\text{CH}_3\text{CCl}_3$ ), is of special importance regarding RFs, not because of its  
54 small RF (see Table 2.1, Figure 2.6), but because this gas is widely used to estimate concentrations of OH  
55 which is the major sink species for  $\text{CH}_4$ , HFCs, and HCFCs and a major production mechanism for sulfate,  
56 nitrate and some organic aerosols as discussed in Section 2.3.5. The global atmospheric methyl chloroform  
57 concentration rose steadily from 1978 to reach a maximum in 1992 (Prinn et al., 2001; Montzka et al., 2003).

1 Since then concentrations have decreased rapidly, driven by a relatively short lifetime of 4.9 years and  
2 phase-out under the Montreal Protocol, to levels in 2003 less than 20% of the levels when AGAGE  
3 measurements peaked in 1992 (Prinn et al., 2005a). Emissions of  $\text{CH}_3\text{CCl}_3$  determined from industry data  
4 (McCulloch and Midgley, 2001) may be too small in recent years. The 2000–2003 emissions for Europe  
5 estimated using surface observations (Reimann et al., 2005), show that 1.2–2.3  $\text{Gg yr}^{-1}$  need to be added over  
6 this 4-year period to the above industry estimates for Europe. Estimates of European emissions in 2000  
7 exceeding 20 Gg (Krol et al., 2003) are not supported by analyses of the above extensive surface data  
8 (Reimann et al., 2005). Emissions for the USA have been estimated by Li et al. (2005). From multi year  
9 measurements they estimate 2001–2002 USA emissions of 2.2  $\text{Gg yr}^{-1}$  (or about half of those estimated from  
10 more temporally but less geographically limited measurements by Millet and Goldstein, 2004), and that  
11 1996–1998 USA emissions may be underestimated by on average about 9.0  $\text{Gg yr}^{-1}$  over this 3-year period.  
12 East Asian emissions deduced from aircraft data in 2001 are about 1.7 Gg above industry data (Palmer et al.,  
13 2003; see also Yokouchi et al., 2005) while recent Australian and Russian emissions are negligible (Prinn et  
14 al., 2001; Hurst et al., 2004).

15  
16 Carbon tetrachloride ( $\text{CCl}_4$ ) is the second most rapidly decreasing atmospheric chlorocarbon after  $\text{CH}_3\text{CCl}_3$ .  
17 Levels peaked in early 1990 and have decreased approximately linearly since then (Figure 2.7). Its major use  
18 was as a feedstock for CFC manufacturing. The fact that (unlike  $\text{CH}_3\text{CCl}_3$ ) a significant interhemispheric  
19  $\text{CCl}_4$  gradient still exists in 2005, in spite of its moderately long lifetime of 20–30 years, results from a  
20 persistence of significant northern hemispheric emissions.

21  
22 [INSERT FIGURE 2.7 HERE]

23  
24 HCFCs of industrial importance have lifetimes in the range of 1.3–20 years. Global and regional emissions  
25 of the CFCs and HCFCs have been derived from observed concentrations and can be used to check emission  
26 inventory estimates. Montzka et al. (2003) and IPCC/TEAP (2005) concluded that global emissions of  
27 HCFC-22 have risen steadily over the period 1975–2000 whilst those of HCFC-141b and HCFC-142b  
28 started increasing quickly in the early 1990s and then began to decrease after 2000.

29  
30 To provide a direct comparison of the effects on global warming due to the annual changes in each of the  
31 non- $\text{CO}_2$  greenhouse gases (discussed in Sections 2.3.2, 2.3.3 and 2.3.4) relative to  $\text{CO}_2$ , Figure 2.7 shows  
32 these annual changes in atmospheric mass multiplied by the GWP (100 year horizon) for each gas (e.g.,  
33 Prinn, 2004). By expressing them in this way, the observed changes in all non- $\text{CO}_2$  gases in GtC equivalents  
34 and the significant roles of  $\text{CH}_4$ ,  $\text{N}_2\text{O}$ , and many halocarbons are very evident. This highlights the  
35 importance of considering the full suite of greenhouse gases for RF calculations.

### 36 37 2.3.5 Trends in the Hydroxyl Free Radical (OH)

38  
39 The hydroxyl free radical (OH) is the major oxidizing chemical in the atmosphere destroying about 3.7 Gt of  
40 trace gases, including  $\text{CH}_4$  and all HFCs and HCFCs, each year (Ehhalt, 1999). It therefore has a very  
41 significant role in limiting the LLGHG RF. IPCC/TEAP (2005) concluded that the OH concentration might  
42 change in the 21st century by –18 to +5% depending on the emission scenario. The large-scale  
43 concentrations and long-term trends in OH can be inferred indirectly using global measurements of trace  
44 gases whose emissions are well known and whose primary sink is OH. The best trace gas used to date for  
45 this purpose is  $\text{CH}_3\text{CCl}_3$  whose long-term measurements are reviewed in Section 2.3.4. Other gases which  
46 are useful OH indicators include  $^{14}\text{CO}$ , which is produced primarily by cosmic rays (Lowe and Allan, 2002).  
47 While the accuracy of the  $^{14}\text{CO}$  cosmic ray and other  $^{14}\text{CO}$  source estimates and also the frequency and  
48 spatial coverage of its measurements do not match those for  $\text{CH}_3\text{CCl}_3$ , the  $^{14}\text{CO}$  lifetime (2 months) is much  
49 shorter than that of  $\text{CH}_3\text{CCl}_3$  (4.9 years). As a result  $^{14}\text{CO}$  provides estimates of average concentrations of  
50 OH that are more regional, and is capable of resolving shorter time scales than those estimated from  
51  $\text{CH}_3\text{CCl}_3$ . The  $^{14}\text{CO}$  source variability is better defined than its absolute magnitude so it is better for inferring  
52 relative rather than absolute trends. Another useful gas is the industrial chemical HCFC-22. It yields OH  
53 concentrations similar to those derived from  $\text{CH}_3\text{CCl}_3$  but with less accuracy due to greater uncertainties in  
54 emissions and less extensive measurements (Miller et al., 1998). The industrial gases HFC-134a, HCFC-  
55 141b and HCFC-142b are potentially useful OH estimators but the accuracy of their emission estimates  
56 needs improvement (Huang and Prinn, 2002; O'Doherty et al., 2004).

1 Indirect measurements of OH using CH<sub>3</sub>CCl<sub>3</sub> have established that the globally weighted average OH  
2 concentration in the troposphere is roughly 10<sup>6</sup> radicals cm<sup>-3</sup> (Prinn et al., 2001; Krol and Lelieveld, 2003).  
3 A similar average concentration is derived using <sup>14</sup>CO (Quay et al., 2000) although the spatial weighting here  
4 is different. Note that methods to infer global or hemispheric average OH concentrations may be insensitive  
5 to compensating regional OH changes such as OH increases over continents and decreases over oceans  
6 (Lelieveld et al, 2002). Also, the quoted absolute OH concentrations (but not their relative trends) depend on  
7 the choice of weighting (e.g., Lawrence et al, 2001). While the global average OH concentration appears  
8 fairly well defined by these indirect methods, the temporal trends in OH are more difficult to discern since  
9 they require long-term measurements, optimal inverse methods, and very accurate calibrations, model  
10 transports, and CH<sub>3</sub>CCl<sub>3</sub> emissions data. From AGAGE CH<sub>3</sub>CCl<sub>3</sub> measurements, Prinn et al. (2001) inferred  
11 that global OH levels grew between 1979 and 1989, but then declined between 1989 and 2000, and also  
12 exhibited significant interannual variations. They concluded that these decadal global variations were driven  
13 principally by northern hemispheric OH, with southern hemispheric OH decreasing from 1979–1989 and  
14 staying essentially constant after that. Using the same AGAGE data and identical CH<sub>3</sub>CCl<sub>3</sub> emissions, a 3D  
15 model analysis (Krol and Lelieveld, 2003) supported qualitatively (but not quantitatively) the earlier result  
16 (Prinn et al., 2001) that OH concentrations increased in the 1980s and declined in the 1990s. Prinn et al.  
17 (2001) also estimated the emissions required to provide a zero trend in OH. These required CH<sub>3</sub>CCl<sub>3</sub>  
18 emissions differed substantially from industry estimates by McCulloch and Midgley (2001) particularly for  
19 1996–2000. Krol and Lelieveld (2003) however argued that the combination of possible underestimated  
20 recent emissions, especially the >20 Gg European emissions deduced by Krol et al. (2003), and the recent  
21 decreasing effectiveness of the stratosphere as a sink for tropospheric CH<sub>3</sub>CCl<sub>3</sub>, may be sufficient to yield a  
22 zero deduced OH trend. As discussed in Section 2.3.4, estimates of European emissions by Reimann et al.  
23 (2005) are an order of magnitude less than those of Krol et al. (2003). Also, Prinn et al. (2005a) extend the  
24 OH estimates through 2004 and show that the Prinn et al. (2001) decadal and interannual OH estimates  
25 remain valid even after accounting for the additional recent CH<sub>3</sub>CCl<sub>3</sub> emissions discussed in Section 2.3.4.  
26 They also reconfirm the OH maximum around 1989 and a larger OH minimum around 1998 with OH  
27 concentrations then recovering so that in 2003 they are comparable to those in 1979. They note that the  
28 1997–1999 OH minimum coincides with, and is *likely* caused by, major global wildfires and an intense El  
29 Niño at this time. The 1997 Indonesian fires alone have been estimated to have lowered global late-1997 OH  
30 levels by 6% due to CO enhancements (Duncan et al., 2003).

31  
32 Methyl chloroform is also destroyed in the stratosphere. Because its stratospheric loss frequency is less than  
33 that in the troposphere, the stratosphere becomes a less effective sink for tropospheric CH<sub>3</sub>CCl<sub>3</sub> over time  
34 (Krol and Lelieveld, 2003), and even becomes a small source to the troposphere beginning in 1999 in the  
35 reference case in the Prinn et al. (2001; 2005a) model. Loss to the ocean has usually been considered  
36 irreversible, and its rates and uncertainties have been obtained from observations (Yvon-Lewis and Butler,  
37 2002). However, Wennberg et al. (2004) have recently proposed that the polar oceans may have effectively  
38 stored methyl chloroform during the pre-1992 years when its atmospheric levels were rising, but began re-  
39 emitting it in the subsequent years, thus lessening the overall oceanic sink. Prinn et al. (2005a) tried both  
40 approaches and found that their inferred interannual and decadal OH variations were present using either  
41 formulation, but inferred OH was lower in the pre-1992 years and higher after that using the Wennberg et al.  
42 (2004) formulation.

43  
44 More recently, Bousquet et al. (2005) have used an inverse method with a 3D model and methyl chloroform  
45 measurements and concluded that substantial year-to-year variations occurred in global-average OH  
46 concentrations between 1980 and 2000. This conclusion was previously reached by Prinn et al. (2001), but  
47 subsequently challenged by Krol and Lelieveld (2003) who argued that these variations are caused by model  
48 shortcomings and that models need, in particular, to include observationally-based, interannually-varying  
49 meteorology to provide accurate annual OH estimates. Neither the 2D Prinn et al. (2001) nor the 3D Krol et  
50 al. (2003) inversion models used interannually-varying circulation. However, the Bousquet et al. (2005)  
51 analysis, which uses observationally-based meteorology and estimates OH on monthly time scales, yields  
52 interannual OH variations that agree very well with the Prinn et al. (2001) and equivalent Krol and Lelieveld  
53 (2003) estimates (see Figure 2.8). However, when Bousquet et al. (2005) estimated both OH concentrations  
54 and methyl chloroform emissions (constrained by their uncertainties as reported by McCulloch and Midgley,  
55 2001), the OH variations are reduced by 65% (dashed line in Figure 2.8). The error bars on the Prinn et al.  
56 (2001, 2005a) OH estimates, which account for these emission uncertainties using Monte Carlo ensembles of  
57 inversions, also easily allow such a reduction in OH variability (thin vertical bars in Figure 2.8). This implies

1 that these interannual OH variations are real, but only their phasing and not their amplitude, is well defined.  
2 Bousquet et al. (2005) also deduced that OH in the southern hemisphere shows a zero to small negative trend  
3 in qualitative agreement with Prinn et al. (2001). Short-term variations in OH have also recently been  
4 deduced by Manning et al. (2005) using 13 years of  $^{14}\text{CO}$  measurements in New Zealand and Antarctica.  
5 They find no significant long-term trend between 1989 and 2003 in southern hemispheric OH but provide  
6 evidence for recurring multi-month OH variations of around 10%. They also deduce even larger (20%) OH  
7 decreases in 1991 and 1997, perhaps triggered by the 1991 Pinatubo eruption and the 1997 Indonesian fires.  
8 The similarity of many of these results to those from  $\text{CH}_3\text{CCl}_3$  discussed above is very important, given the  
9 independence of the two approaches.

10  
11 [INSERT FIGURE 2.8 HERE]

12  
13 RF calculations of the LLGHGs are calculated from observed trends in the LLGHG concentrations and  
14 therefore OH concentrations do not directly affect them. Nevertheless OH trends are needed to quantify  
15 LLGHG budgets (Chapter 7: Section 7.4) and for understanding future trends in the LLGHGs and  
16 tropospheric ozone.

### 17 18 **2.3.6 Ozone**

19 In the TAR, separate estimates for RF due to change in tropospheric ozone and stratospheric ozone were  
20 given. Stratospheric ozone RF was derived from observations of ozone change from roughly 1979-1998.  
21 Tropospheric ozone RF was based on chemical model results employing changes in precursor hydrocarbons,  
22  $\text{CO}$ , and  $\text{NO}_x$ . Over the satellite era (since approximately 1980) stratospheric ozone trends have been  
23 primarily caused by the Montreal Protocol gases, and in Ramaswamy et al. (2001) the stratospheric ozone  
24 RF was implicitly attributed to these gases. Studies since then have investigated a number of possible causes  
25 of ozone change in the stratosphere and troposphere and the attribution of ozone trends to a given precursor  
26 is less clear. Nevertheless, stratospheric ozone and tropospheric ozone RFs are still treated separately in this  
27 report. However, the RFs are more associated with the vertical location of the ozone change than they are  
28 with the agent(s) responsible for the change.

#### 29 30 **2.3.6.1 Stratospheric Ozone**

31  
32 The TAR reported that ozone depletion in the stratosphere had caused a negative RF of  $-0.15 \text{ W m}^{-2}$  as a  
33 best estimate over the period since 1750. A number of recent reports have assessed changes in stratospheric  
34 ozone and the research into its causes: Chapters 3 and 4 of the 2002 WMO Scientific Assessment of Ozone  
35 Depletion (WMO, 2003) and Chapter 1 of IPCC/TEAP (2005). This section summarizes the material from  
36 these reports and updates the key results using more recent research.

37  
38 Global ozone amounts decreased between the late 1970s and early 1990s, with the lowest values occurring  
39 during 1992–1993 (roughly 6% below the 1964–1980 average), and slightly increasing values thereafter.  
40 Global ozone for the period 2000 to 2003 was approximately 4% below the 1964–1980 average values.  
41 Whether or not recently observed changes in ozone trends (Newchurch et al., 2003; Weatherhead and  
42 Andersen, 2006) are already indicative of recovery of the global ozone layer is not yet clear and requires  
43 more detailed attribution of the drivers of the changes (Steinbrecht et al., 2004a (see also comment and reply:  
44 Cunnold et al., 2004 and Steinbrecht et al., 2004b); Hadjinicolaou et al., 2005; Krizan and Lastovicka, 2005;  
45 Weatherhead and Andersen, 2006). The largest ozone changes since 1980 have occurred during the late  
46 winter and spring over Antarctica where average total column ozone in September and October is about 40–  
47 50% below pre-1980 values (WMO, 2003). Ozone decreases over the Arctic have been less severe than those  
48 over the Antarctic due to higher temperature in the lower stratosphere and thus less polar stratospheric clouds  
49 to cause the chemical destruction. Arctic stratospheric ozone levels are more variable due to interannual  
50 variability in chemical loss and transport.

51  
52 The temporally and seasonally non-uniform nature of stratospheric ozone trends has important implications  
53 for the resulting RF. Global ozone decreases come primarily from changes in the lower stratospheric  
54 extratropics. Total column ozone changes over the midlatitudes of the southern hemisphere are significantly  
55 larger than over the midlatitudes of the northern hemisphere. Averaged over the period 2000–2003, southern  
56 hemisphere values are 6% below pre-1980 values, while northern hemisphere values are 3% lower. There is  
57 also significant seasonality in the northern hemisphere ozone changes with 4% decreases in winter to spring

1 and 2% decreases in summer, while long-term southern hemisphere changes are roughly 6% year round  
2 (WMO, 2003). Southern hemisphere mid-latitude ozone shows significant decreases during the mid-1980s  
3 and essentially no response to the effects of the Mt. Pinatubo volcanic eruption in June 1991; both of these  
4 features remain unexplained. Pyle et al. (2005) and Chipperfield et al. (2003) assess several studies that show  
5 that a substantial fraction (roughly 30%) of northern hemisphere midlatitude ozone trends are not directly  
6 attributable to anthropogenic chemistry, but are related to dynamical effects, such as tropopause height  
7 changes. These dynamical effects are *likely* to have contributed a larger fraction of the ozone RF in the  
8 northern hemisphere midlatitudes. The only study to assess this finds that 50% of the RF related to  
9 stratospheric ozone changes between 20°N–60°N over the period 1970–1997 is attributable to dynamics  
10 (Forster and Tourpali, 2001). These dynamical changes may well have an anthropogenic origin and could  
11 even be partly caused by stratospheric ozone changes themselves through lower stratospheric temperature  
12 changes (Chipperfield et al., 2003; Santer et al., 2004), but are not directly related to chemical ozone loss.

13  
14 At the time of writing no study has utilised ozone trend observations after 1998 to update the RF values  
15 presented in the Ramaswamy et al. (2001). However, Hansen et al. (2005) repeated the RF calculation based  
16 on the same trend dataset employed by studies assessed in Ramaswamy et al. (2001) and found an RF of  
17 roughly  $-0.06 \text{ W m}^{-2}$ . A considerably stronger RF of  $-0.2 \pm 0.1 \text{ W m}^{-2}$  previously estimated by the same  
18 group affected the Ramaswamy et al. (2001) assessment. The two other studies assessed in Ramaswamy et  
19 al. (2001) using similar trend datasets found RFs of  $-0.01 \text{ W m}^{-2}$  and  $-0.10 \text{ W m}^{-2}$ . Using the three estimates  
20 gives a revision of the observational-based RF for 1979–1998 to about  $-0.05 \pm 0.05 \text{ W m}^{-2}$ .

21  
22 Gauss et al. (2006) compared results from six chemical transport models that included changes in ozone  
23 precursors to simulate both the increase in the ozone in the troposphere and the ozone reduction in the  
24 stratosphere over the industrial era. The 1850–2000 annually averaged global mean stratospheric ozone  
25 column reduction for these models ranged between 14 DU and 29 DU. The overall pattern of the ozone  
26 changes from the models were similar but the magnitude of the ozone changes differed. The models show a  
27 reduction in the ozone at high latitudes, ranging from around 20 to 40% in the southern hemisphere and  
28 smaller changes in the northern hemisphere. All models have a maximum ozone reduction around 15 km at  
29 high latitudes in the southern hemisphere. Differences between the models were also found in the tropics,  
30 with some models giving around 10% increase in the lower stratosphere, whereas other models show  
31 decreases. These differences were especially related to the altitude where the ozone trend switched from an  
32 increase in the troposphere to a decrease in the stratosphere ranging from close to the tropopause to around  
33 27 km. Several studies have shown that ozone changes in the tropical lower stratosphere are very important  
34 for the magnitude and sign of the ozone RF (Ramaswamy et al., 2001). The resulting stratospheric ozone RF  
35 ranged between  $-0.12 \text{ W m}^{-2}$  and  $+0.07 \text{ W m}^{-2}$ . Note that the models with either a small negative or a  
36 positive RF also had a small increase in tropical lower stratospheric ozone, resulting from increases in  
37 tropospheric ozone precursors; most of this increase would have occurred before the time of stratospheric  
38 ozone destruction by the Montreal Protocol gases. These RF calculations also did not include any negative  
39 RF that may have resulted from stratospheric water vapour increases. It has been suggested (Shindell and  
40 Faluvegi, 2002) that stratospheric ozone during 1957 to 1975 was lower by about 7 DU relative to the first  
41 half of the 20th century as a result of possible stratospheric water vapour increases; however, these long-term  
42 increases in stratospheric water vapour are uncertain (see Section 2.3.7 and Chapter 3, Section 3.4).

43  
44 The stratospheric ozone RF is assessed to be  $-0.05 \pm 0.10 \text{ W m}^{-2}$  between preindustrial times and 2005. The  
45 best-estimate is from the observationally based 1979–1998 RF of  $-0.05 \pm 0.05 \text{ W m}^{-2}$ , with the uncertainty  
46 range increased to take into account ozone change prior to 1979, using the model results of Gauss et al.  
47 (2006) as a guide. Note that this estimate takes into account causes of stratospheric ozone change in addition  
48 to those due to the Montreal Protocol gases. The level of scientific understanding is *medium*, unchanged  
49 since the TAR (Table 2.11).

### 50 51 2.3.6.2 Tropospheric Ozone

52  
53 The TAR report identified large regional differences in observed trends in tropospheric ozone from  
54 ozonesondes and surface observations. The TAR estimate of RF from tropospheric ozone was  $0.35 \pm 0.15 \text{ W m}^{-2}$ .  
55 Due to limited spatial and temporal coverage of observations of tropospheric ozone, the RF estimate is  
56 based on model simulations. In the TAR, the models considered only changes in the tropospheric  
57 photochemical system, driven by estimated emission changes ( $\text{NO}_x$ , CO, NMVOC, and  $\text{CH}_4$ ) since

1 preindustrial times. Since the TAR there have been major improvements in models. The new-generation  
2 models include several chemical tracer models (CTM) which couple stratospheric and tropospheric  
3 chemistry, as well as GCMs with online chemistry (both tropospheric and stratospheric). While the TAR  
4 simulations did not consider changes in ozone within the troposphere caused by reduced influx of ozone  
5 from the stratosphere (due to ozone depletion in the stratosphere), the new models include this process  
6 (Gauss et al., 2006). This advancement in modelling capabilities and the need to be consistent with how the  
7 RF due to changes in stratospheric ozone is derived (based on observed ozone changes) have led to a change  
8 in the definition of RF due to tropospheric ozone compared with that in the TAR. Changes in tropospheric  
9 ozone due to changes in transport of ozone across the tropopause, which are in turn caused by changes in  
10 stratospheric ozone, are now included.

11  
12 Trends in anthropogenic emissions of ozone precursors for the period 1990–2000 have been compiled by the  
13 Emission Database for Global Atmospheric Research (EDGAR) consortium (Olivier and Berdowski, 2001  
14 updated). For specific regions there is significant variability over the period due to variations in the  
15 emissions from open biomass burning sources. For all components ( $\text{NO}_x$ , CO and VOCs) industrialized  
16 regions like USA and OECD Europe show reductions in the emissions, while regions dominated by  
17 developing countries show significant growth in the emissions. Recently, the tropospheric burden of CO and  
18  $\text{NO}_2$  have been estimated from satellite observations (Edwards et al., 2004; Richter et al., 2005), providing  
19 much needed data for model evaluation and very valuable constraints for emission estimates.

20  
21 Assessment of long-term trends in tropospheric ozone is difficult due to the scarcity of representative  
22 observing sites with long records. The long-term tropospheric ozone trends vary both in terms of sign and  
23 magnitude and in the possible causes for the change (Oltmans et al., 2006). Trends in tropospheric ozone at  
24 northern mid- and high latitudes have been estimated based on ozonesonde data by WMO (2003), Naja et al.  
25 (2003), Naja and Akimoto (2004), Tarasick et al. (2005), and Oltmans et al. (2006). Over Europe, ozone in  
26 the free troposphere increased from the early 20<sup>th</sup> century until the late 1980s, while in the later period the  
27 trend has levelled off or been slightly negative. Naja and Akimoto (2004) analysed 33 years of ozonesonde  
28 data from Japanese stations showing an increase in ozone in the lower troposphere (750–550 hPa) between  
29 the periods 1970–1985 and 1986–2002 of 12–15% at Sapporo and Tsukuba (43° and 36°N) and 35% at  
30 Kagoshima (32°N). Trajectory analysis indicates that the more southerly located station Kagoshima is  
31 significantly more influenced by air originating over China, while Sapporo and Tsukuba are more influenced  
32 by air from Eurasia. At Naha (26°N) a positive trend (5% decade<sup>-1</sup>) is found between 700 and 300 hPa (1990-  
33 2004), while between the surface and 700 hPa a slightly negative trend is observed (Oltmans et al., 2006).  
34 Ozonesondes from Canadian stations show negative trends of tropospheric ozone between 1980 and 1990,  
35 and a rebound with positive trends during 1991–2001 (Tarasick et al. 2005). Analysis of stratosphere-  
36 troposphere exchange (STE) processes indicates that the rebound during the 1990s may be partly a result of  
37 small changes in atmospheric circulation. Trends are also derived from surface observations. Jaffe et al.  
38 (2003) derived a positive trend 1.4% yr<sup>-1</sup> between 1988 and 2003 using measurements from Lassen Volcanic  
39 Park in California (1750 m above sea level) consistent with the trend derived by comparing two aircraft  
40 campaigns (Parrish et al., 2004). A number of other sites show, however, insignificant changes over the  
41 United States over the last 15 years (Oltmans et al. 2006). Over Europe and North America, observations  
42 from Whiteface Mountain, Wallops Island, Hohenpeisenberg, Zugspitze, and Mace Head (flow from the  
43 European sector) show small trends or reductions during summer, while there is an increase during winter  
44 (Oltmans et al., 2006). These observations are consistent with reduced  $\text{NO}_x$  emissions (Jonson et al., 2005).  
45 North Atlantic stations (Mace Head, Izana, and Bermuda) indicate increased ozone (Oltmans et al., 2006).  
46 Over the North Atlantic (40–60°N) measurements from ships (Lelieveld et al., 2004) show insignificant  
47 trends in ozone, however, at Mace Head a positive trend of  $0.49 \pm 0.19$  ppb yr<sup>-1</sup> for the period 1987–2003 is  
48 found, with the largest contribution from air coming from the Atlantic sector (Simmonds et al., 2004).

49  
50 In the tropics very few long-term ozonesonde measurements are available. At Irene in South Africa (26°S),  
51 Diab et al. (2004) found an increase between the 1990–1994 and 1998–2002 periods of about 10 ppbv close  
52 to the surface (except in summer) and in the upper troposphere during winter. Thompson et al. (2001) found,  
53 based on TOMS satellite data, no significant trend during 1979–1992. More recent observations (1994–2003,  
54 in-situ data from the MOZAIC program) show significant trends in free-tropospheric ozone (7.7–11.3 km  
55 altitude) in the tropics:  $1.12 \pm 0.05$  ppbv yr<sup>-1</sup> and  $1.03 \pm 0.08$  ppbv yr<sup>-1</sup> in the NH tropics and SH tropics,  
56 respectively (Bortz and Prather, 2006). Ozonesonde measurements over the South West Pacific indicate an

1 increased frequency of near-zero ozone in the upper troposphere, suggesting a link to an increased frequency  
2 of deep convection there since the 1980s (Solomon et al., 2005).

3  
4 At southern mid-latitudes surface observations from Cape Point, Cape Grim, Atlantic Ocean (from ship) and  
5 from sondes at Lauder (850–700 hPa) show positive trends in ozone concentrations, in particular during the  
6 biomass burning season in the SH (Oltmans et al., 2006). However, the trend is not accompanied by a similar  
7 trend in CO, as expected if biomass burning had increased. The increase is largest at Cape Point, reaching  
8 20%/decade (in September). At Lauder the increase is confined to the lower troposphere.

9  
10 Changes in tropospheric ozone and the corresponding RF have been estimated in a number of recent model  
11 studies (Hauglustaine and Brasseur, 2001; Mickley et al., 2001; Shindell et al., 2003a; Mickley et al., 2004;  
12 Wong et al., 2004; Liao and Seinfeld, 2005; Shindell et al., 2005). In addition, a multi-model experiment  
13 including 10 global models was organized through the ACCENT Network (Gauss et al., 2006). Four of the  
14 ten ACCENT models have detailed stratospheric chemistry. The adjusted RF for all models was calculated  
15 by the same radiative transfer model. The normalized adjusted RF for the ACCENT models was  $0.032 \pm$   
16  $0.006 \text{ W m}^{-2} \text{ DU}^{-1}$ , which is significantly lower than the TAR estimate of  $0.042 \text{ W m}^{-2} \text{ DU}^{-1}$ .

17  
18 The simulated RFs for tropospheric ozone increases since 1750 are shown in Figure 2.9. Most of the  
19 calculations used the same set of assumptions about preindustrial emissions (zero anthropogenic emissions  
20 and biomass burning sources reduced by 90%). Emissions of NO<sub>x</sub> from soils and biogenic hydrocarbons  
21 were generally assumed to be natural and were thus not changed (cf. Chapter 7, Section 7.4). In one study  
22 (Hauglustaine and Brasseur, 2001) preindustrial NO<sub>x</sub> emissions from soils were reduced based on changes in  
23 the use of fertilizers. Six of the ACCENT models also made coupled climate-chemistry simulations  
24 including climate change since pre-industrial times. The difference between the RF in the coupled climate-  
25 chemistry and the chemistry only simulations, which indicate the possible climate feedback on tropospheric  
26 ozone, was positive in all models but generally small (Figure 2.9).

27  
28 [INSERT FIGURE 2.9 HERE]

29  
30 A general feature of the models is their inability to reproduce the low ozone concentrations indicated by the  
31 very uncertain semi-quantitative observations (e.g., Pavelin et al., 1999) during the late 19th century.  
32 Mickley et al. (2001) tuned their model by reducing pre-industrial lightning and soil sources of NO<sub>x</sub> and  
33 increasing natural non-methane volatile organic compound (NMVOC) emissions to obtain close agreement  
34 with the observations. The ozone RF then increased by 50–80% compared to their standard calculations.  
35 However, there are still several aspects of the early observations that were not captured by the tuned model.

36  
37 The best estimate for the RF of tropospheric ozone increases is  $0.35 \text{ W m}^{-2}$ , taken as the median of the RF  
38 values in Figure 2.9 (adjusted and non climate change values only, i.e., the red bars). The best estimate is  
39 unchanged from the TAR. The uncertainties in the estimated RF by tropospheric ozone, originates from two  
40 factors: The models used (CTM/GCM model formulation, radiative transfer models), and the potential  
41 overestimation of preindustrial ozone levels in the models. The 5–95% confidence interval, assumed to be  
42 represented by the range of the results in Figure 2.9, is  $0.25\text{--}0.65 \text{ W m}^{-2}$ . A *medium* level of scientific  
43 understanding is adopted, also unchanged from the TAR (Table 2.11).

### 44 45 2.3.7 Stratospheric Water Vapour

46  
47 The TAR noted that several studies had indicated long-term increases in stratospheric water vapour and  
48 acknowledged that these trends would contribute a significant radiative impact. However, it only considered  
49 the stratospheric water vapour increase expected from methane increases as an RF, and this was estimated to  
50 contribute 2 to 5% of the total methane RF (about  $0.02 \text{ W m}^{-2}$ ).

51  
52 Chapter 3 (Section 3.4) discusses the evidence for stratospheric water vapour trends and presents the current  
53 understanding of their possible causes. There are now 14 years of global stratospheric water vapour  
54 measurements from HALOE and continued balloon-based measurements (since 1980) at Boulder, Colorado.  
55 There is some evidence of a sustained long-term increase in stratospheric water vapour of around  $0.05 \text{ ppm}$   
56  $\text{year}^{-1}$  from 1980 until roughly 2000. As well as CH<sub>4</sub> increases, several other indirect forcing mechanisms  
57 have been proposed, including: a) volcanic eruptions (Considine et al., 2001; Joshi and Shine, 2003) b)

1 biomass burning aerosol (Sherwood, 2002); c) tropospheric SO<sub>2</sub> (Notholt et al., 2005) and d) changes to  
2 methane oxidation rates from changes in stratospheric chlorine, ozone and OH (Rockmann et al., 2004).  
3 These are mechanisms which can be linked to an external forcing agent. Other proposed mechanisms are  
4 more associated with climate feedbacks and are related to changes in tropopause temperatures or circulation  
5 (Stuber et al., 2001b; Fueglistaler et al., 2004). From these studies there is little quantification of the  
6 stratospheric water vapour change attributable to different causes. It is also *likely* that different mechanisms  
7 are affecting water vapour trends at different altitudes.

8  
9 Since the TAR several further calculations of the radiative balance change due to changes in stratospheric  
10 water vapour have been performed (Forster and Shine, 1999; Oinas et al., 2001; Shindell, 2001; Smith et al.,  
11 2001; Forster and Shine, 2002). Smith et al. (2001) estimated a 0.12 to 0.2 W m<sup>-2</sup> per decade range for the  
12 RF from the change in stratospheric water vapour, using HALOE satellite data. Shindell (2001) estimated an  
13 RF of about 0.2 W m<sup>-2</sup> in a period of two decades, using a GCM to estimate the increase in water vapour in  
14 the stratosphere from oxidation of CH<sub>4</sub> and including climate feedback changes associated with an increase  
15 in greenhouse gases. Forster and Shine (2002) used a constant 0.05 ppm yr<sup>-1</sup> trend of water vapour at  
16 pressures of 100–10 hPa and estimated the RF to be 0.29 W m<sup>-2</sup> for 1980 to 2000. GCM radiation codes can  
17 have a factor of two uncertainty in their modelling of this RF (Oinas et al., 2001). For the purposes of this  
18 chapter the above RF estimates are not readily attributable to forcing agent(s) and uncertainty as to the  
19 causes of the observed change preclude all but the component due to CH<sub>4</sub> increases being considered a  
20 forcing. Two related chemical transport model studies have calculated the RF associated with increases in  
21 CH<sub>4</sub> since preindustrial times (Hansen and Sato, 2001 and Hansen et al., 2005), but no dynamical feedbacks  
22 were included in these estimates. Hansen et al. (2005) estimate an RF of 0.07 ± 0.01 W m<sup>-2</sup> for the  
23 stratospheric water vapour changes over 1750–2000, which is at least a factor of three larger than the TAR  
24 value. The RF from direct injection of water vapour by aircraft is believed to be an order of magnitude  
25 smaller than this at about 0.002 W m<sup>-2</sup> (IPCC, 1999). There has been little trend in CH<sub>4</sub> concentration since  
26 2000 (see Section 2.3.2); therefore the best estimate of the stratospheric water vapour RF from CH<sub>4</sub>  
27 oxidation (0.07 W m<sup>-2</sup>) is based on the Hansen et al. (2005) calculation. The 90% confidence range is  
28 estimated as ±0.05 W m<sup>-2</sup>, from the range of the RF studies which included other effects. There is a *low* level  
29 of scientific understanding in this estimate, as there is only a partial understanding of the vertical profile of  
30 methane-induced stratospheric water vapour change (Table 2.11). Other human causes of stratospheric water  
31 vapour change are unquantified and have a *very low* level of scientific understanding.

### 32 33 **2.3.8 Observations of Long-Lived Greenhouse Gas Radiative Effects**

34  
35 Observations of the clear-sky radiation emerging at the top of atmosphere and surface have been conducted.  
36 Such observations, by their nature, do not measure RF as defined here. Instead they yield a perspective on  
37 the influence of various species on the transfer of radiation in the atmosphere. Most importantly, the  
38 conditions involved with these observations involve varying thermal and moisture profiles in the atmosphere  
39 such that they do not conform to the conditions underlying the RF definition (see Section 2.2). There is a  
40 more comprehensive discussion of observations of the Earth's radiative balance in Chapter 3 (Section 3.4).

41  
42 Harries et al. (2001) analyzed spectra of the outgoing longwave radiation as measured by two satellites in  
43 1970 and 1997 over the tropical Pacific Ocean. The reduced brightness temperature observed in the spectral  
44 regions of many of the greenhouse gases is experimental evidence for an increase in the Earth's greenhouse  
45 effect. In particular the spectral signatures were large for CO<sub>2</sub> and CH<sub>4</sub>. The halocarbons, with their large  
46 change between 1970 and 1997 also had an impact on the brightness temperature. Philipona et al. (2004)  
47 found an increase in the measured longwave downward radiation at the surface over the period from 1995 to  
48 2002 at eight stations over the central Alps. A significant increase in the clear-sky longwave downward flux  
49 was found to be due to an enhanced greenhouse effect after combining the measurements with model  
50 calculations to estimate the contribution from increases in temperature and humidity. While both types of  
51 observations attest to the radiative influences of the gases, they should not be interpreted as having a direct  
52 linkage to the value of RFs in Section 2.3.

## 53 54 **2.4 Aerosols**

### 55 56 **2.4.1 Introduction and Summary of the Third Assessment Report**

1 The TAR categorised aerosol RFs into direct and indirect effects. The direct effect is the mechanism by  
2 which aerosols scatter and absorb shortwave and longwave radiation, thereby altering the radiative balance  
3 of the Earth-atmosphere system. Sulphate, fossil-fuel organic carbon, fossil-fuel black carbon, biomass  
4 burning, and mineral dust aerosols were all identified as having a significant anthropogenic component and  
5 exerting a significant direct RF. Key parameters for determining the direct RF are the aerosol optical  
6 properties (the single scattering albedo,  $\omega_0$ , specific extinction coefficient,  $k_e$ , and scattering phase function)  
7 which vary as a function of wavelength and relative humidity, and the atmospheric loading and geographic  
8 distribution of the aerosols in the horizontal and vertical which vary as a function of time (e.g., Haywood and  
9 Boucher, 2000; Penner et al., 2001; Ramaswamy et al., 2001). Scattering aerosols exert a net negative direct  
10 RF, while partially absorbing aerosols may exert a negative top-of-the-atmosphere (TOA) direct RF over  
11 dark surfaces such as oceans or dark forest surfaces, and a positive TOA RF over bright surfaces such as  
12 desert, snow/ice or if the aerosol is above cloud (e.g., Chylek and Wong, 1995; Haywood and Shine, 1995).  
13 Both positive and negative TOA direct RF mechanisms reduce the shortwave irradiance at the surface. The  
14 longwave direct RF is only substantial if the aerosol particles are large and occur in considerable  
15 concentrations at higher altitudes (e.g., Tegen et al., 1996). The direct RF due to tropospheric aerosols is  
16 most frequently derived at TOA rather than at the tropopause because shortwave radiative transfer  
17 calculations have shown a negligible difference between the two (e.g., Haywood and Shine, 1997 and  
18 Section 2.2). The *surface forcing* will be approximately the same as the direct RF at the TOA for scattering  
19 aerosols, but for partially absorbing aerosols the *surface forcing* may be many times stronger than the TOA  
20 direct RF (e.g., Ramanathan et al., 2001b and references therein).

21  
22 The indirect effect is the mechanism by which aerosols modify the microphysical and hence the radiative  
23 properties, amount and lifetime of clouds (Figure 2.10). Key parameters for determining the indirect effect  
24 are the effectiveness of an aerosol particle to act as a cloud condensation nucleus (CCN), which is a function  
25 of the size, chemical composition, mixing state and ambient environment (e.g., Penner et al., 2001). The  
26 microphysically induced effect on the cloud droplet number concentration and hence the cloud droplet size,  
27 with the liquid water content held fixed has been called the *first indirect effect* (e.g., Ramaswamy et al.,  
28 2001), the *cloud albedo effect* (e.g., Lohmann and Feichter, 2005), or the *Twomey effect* (e.g., Twomey,  
29 1977). The microphysically induced effect on the liquid water content, cloud height, and lifetime of clouds  
30 has been called the *second indirect effect* (e.g., Ramaswamy et al., 2001), the *cloud lifetime effect* (e.g.,  
31 Lohmann and Feichter, 2005) or the *Albrecht effect* (e.g., Albrecht, 1989). The TAR split the indirect effect  
32 into the first indirect effect), and the second indirect effect. Throughout this report, these effects are denoted  
33 as “*cloud albedo effect*” and “*cloud lifetime effect*”, respectively as these terms are more descriptive of the  
34 microphysical processes that occur. The cloud albedo effect was considered in the TAR to be an RF because  
35 global model calculations could be performed to describe the influence of increased aerosol concentration on  
36 the cloud optical properties while holding the liquid water content of the cloud fixed, i.e., in an entirely  
37 diagnostic manner where feedback mechanisms do not occur. The TAR considered the cloud albedo effect to  
38 be a key uncertainty in the RF of climate but did not assign a best estimate of the RF, and showed a range of  
39 RF between 0 and  $-2 \text{ W m}^{-2}$  in the context of liquid water clouds. The other indirect effects were not  
40 considered to be an RF because, in suppressing drizzle, increasing the cloud height, or the cloud lifetime in  
41 atmospheric models (Figure 2.10), the hydrological cycle is invariably altered, i.e., feedbacks occur (see  
42 Chapter 7, Section 7.5). The TAR also discussed the impact of anthropogenic aerosols in the formation and  
43 modification of the physical and radiative properties of ice clouds (Penner et al., 2001), although  
44 quantification of an RF from this mechanism was not considered appropriate given the host of uncertainties  
45 and unknowns surrounding ice cloud nucleation and physics.

46  
47 [INSERT FIGURE 2.10 HERE]

48  
49 The TAR did not include any assessment of the semi-direct effect (e.g., Hansen et al. 1997; Ackerman et al.,  
50 2000a; Jacobson, 2002; Menon et al., 2003; Cook and Highwood, 2004; Johnson et al., 2004) which is the  
51 mechanism by which absorption of shortwave radiation by tropospheric aerosols leads to heating of the  
52 troposphere which, in turn changes the relative humidity and the stability of the troposphere and thereby  
53 influences cloud formation and lifetime. In this report, the semi-direct effect is not strictly considered an RF  
54 because of modifications to the hydrological cycle, as discussed in Chapter 7, Section 7.5 (see also Sections  
55 2.2, 2.8 and 2.4.5).

1 Since the TAR there have been substantial developments in observations and modelling of tropospheric  
2 aerosols; these are discussed in turn in the following sections.

### 3 4 **2.4.2 *Developments Related to Aerosol Observations***

5  
6 Surface based measurements of in situ aerosol properties such as size distribution, chemical composition,  
7 scattering and absorption continue to be performed at a number of sites, either at long-term monitoring sites,  
8 or specifically as part of intensive field campaigns. These in situ measurements provide essential validation  
9 for global models, e.g., by constraining aerosol concentrations at the surface and by providing high-quality  
10 information about chemical composition and local trends. In addition, they provide key information on  
11 variability on various timescales. Comparisons of in situ measurements against those from global  
12 atmospheric models are complicated by differences in meteorological conditions and because in situ  
13 measurements are representative of conditions mostly at or near the surface while the direct and indirect RF  
14 depend on the aerosol vertical profile. For example, the spatial resolution of global model grid-boxes are  
15 typically a few degrees of latitude and longitude and the time-steps for the atmospheric dynamics and  
16 radiation calculations may be minutes to hours depending on the process to be studied; this poses limitations  
17 when comparing with observations conducted over smaller spatial extent and shorter time duration.

18  
19 Combinations of satellite and surface-based observations provide near-global retrievals of aerosol properties.  
20 These are discussed in this sub-section; the emissions estimates, trends, and in situ measurements of the  
21 physical and optical properties are discussed with respect to their influence on RF in Section 2.4.4. Further  
22 detailed discussions of the recent satellite observations of aerosol properties and a satellite measurement  
23 based assessment of the aerosol direct RF are given by Yu et al. (2006).

#### 24 25 **2.4.2.1 *Satellite Retrievals***

26  
27 Satellite retrievals of aerosol optical depth in cloud-free regions have improved via new-generation sensors  
28 (Kaufman et al., 2002) and an expanded global validation program (Holben et al., 2001). Advanced aerosol  
29 retrieval products such as aerosol fine-mode fraction and effective particle radius have been developed and  
30 offer potential for improving estimates of the aerosol direct radiative effect. Additionally, efforts have been  
31 made to determine the anthropogenic component of aerosol and associated direct RF, as discussed by  
32 Kaufman et al. (2002) and implemented by Bellouin et al. (2005) and Chung et al. (2005). However,  
33 validation programs for these advanced products have yet to be developed and initial assessments indicate  
34 some systematic errors (Levy et al., 2003; Anderson et al., 2005a; Chu et al., 2005), suggesting that the  
35 routine differentiation between natural and anthropogenic aerosols from satellite retrievals remains very  
36 challenging.

##### 37 38 **2.4.2.1.1 *Satellite retrievals of aerosol optical depth, $\tau_{aer}$***

39 Figure 2.11 shows an example of  $\tau_{aer}$  (mid-visible wavelength) retrieved over both land and ocean, together  
40 with geographical positions of aerosol instrumentation. Table 2.2 provides a summary of aerosol data  
41 currently available from satellite instrumentation, together with acronyms for the instruments.  $\tau_{aer}$  from the  
42 MODIS instrument for the January to March 2001 average (Figure 2.11a) clearly differs from that for the  
43 August to October 2001 average (Figure 2.11b) (Kaufman et al., 1997; Tanré et al., 1997). Seasonal  
44 variability in  $\tau_{aer}$  can be seen; biomass burning aerosol is most strongly evident over the Gulf of Guinea in  
45 Figure 2.11a but shifts to southern Africa in Figure 2.11b. Likewise the biomass burning in South America is  
46 most evident in Figure 2.11b. In Figure 2.11a transport of mineral dust from Africa to South America is  
47 discernible while in Figure 2.11b mineral dust is transported over the West Indies and Central America.  
48 Industrial aerosol which consists of a mixture of sulphates, organic and black carbon, nitrates, and industrial  
49 dust is evident over many continental regions of the northern hemisphere. Sea-salt aerosol is visible in  
50 oceanic regions where the wind speed is high (e.g., south of 45°S). The MODIS aerosol algorithm is  
51 currently unable to make routine retrievals over highly reflective surfaces such as deserts, snow cover, ice,  
52 and areas affected by ocean glint, as well as over high latitude regions when the solar insolation is  
53 insufficient.

54  
55 [INSERT TABLE 2.2 HERE]

56  
57 [INSERT FIGURE 2.11 HERE]

1  
2 Early retrievals for estimating  $\tau_{\text{aer}}$  include the single channel retrieval of the AVHRR (e.g., Husar et al.,  
3 1997, Ignatov and Stowe, 2002), and the UV based retrieval from the TOMS (e.g., Torres et al., 2002). A  
4 dual-channel AVHRR retrieval has also been developed (e.g., Mishchenko et al., 1999; Geogdzhayev et al.,  
5 2002). AVHRR retrievals are generally only performed over ocean surfaces where the surface reflectance  
6 characteristics are relatively well known, although retrievals are also possible over dark land surfaces such as  
7 boreal forests and lakes (Soufflet et al., 1997). The TOMS retrieval is essentially independent of surface  
8 reflectance thereby allowing retrievals over both land and ocean (Torres et al., 2002), but is sensitive to the  
9 altitude of the aerosol, and has a relatively low spatial resolution. While these retrievals only use a limited  
10 number of spectral bands and lack sophistication compared to those from dedicated satellite instruments,  
11 they have the advantage of offering continuous long-term data sets (e.g., Geogdzhayev et al., 2002).

12 Early retrievals have been superseded by those from dedicated aerosol instruments (e.g., Kaufman et al.,  
13 2002). POLDER uses a combination of spectral channels (0.44–0.91  $\mu\text{m}$ ) with several viewing angles, and  
14 measures polarization of radiation.  $\tau_{\text{aer}}$  and Ångström exponent,  $\alpha$ , over ocean (Deuzé et al., 2000),  $\tau_{\text{aer}}$  over  
15 land (Deuzé et al., 2001), and the direct radiative effect of aerosols (Boucher and Tanré, 2000; Bellouin et  
16 al., 2003) have all been developed. Algorithms for aerosol retrievals using MODIS have been developed and  
17 validated over both ocean (Tanré et al., 1997) and land surfaces (Kaufman et al., 1997). The uncertainty in  
18 these retrievals of  $\tau_{\text{aer}}$  is necessarily higher over land (Chu et al., 2002) than over oceans (Remer et al., 2002)  
19 owing to uncertainties in land surface reflectance characteristics but can be minimised by careful selection of  
20 the viewing geometry (Chylek et al., 2003). In addition, new algorithms have been developed for  
21 discriminating between sea-salt/dust/biomass burning and industrial pollution over oceans (Bellouin et al.,  
22 2003; Bellouin et al., 2005; Kaufman et al., 2005a) which allow for a more comprehensive comparison  
23 against aerosol models. MISR retrievals have been developed using multiple viewing capability to determine  
24 aerosol parameters over ocean (Kahn et al., 2001) and land surfaces, including highly reflectant surfaces  
25 such as deserts (Martonchik et al., 2004). Five typical aerosol climatologies, each containing four aerosol  
26 components, are used in the retrievals, and the optimum radiance signature is determined for nine viewing  
27 geometries and two different radiances. The results have been validated against those from AERONET  
28 (Aerosol RObotic NETwork; see Section 2.4.3). ATSR and ATSR-2 retrievals (Veefkind et al., 1998;  
29 Holzer-Popp et al., 2002) use a relatively wide spectral range (0.56–1.65  $\mu\text{m}$ ), and two viewing directions  
30 and aerosol climatologies from the Optical Parameters of Aerosols and Clouds (OPAC) database (Hess et al.,  
31 1998) to make  $\tau_{\text{aer}}$  retrievals over both ocean and land (Robles-Gonzalez et al., 2000). The OCTS retrieval  
32 has a similar basis to the dual wavelength retrieval from AVHRR and uses wavelengths over the range 0.41–  
33 0.86  $\mu\text{m}$  to derive  $\tau_{\text{aer}}$  and  $\alpha$  over oceans (e.g., Higurashi et al., 2000) using a bi-modal aerosol size  
34 distribution. SeaWiFs uses 0.765  $\mu\text{m}$  and 0.856  $\mu\text{m}$  radiances to provide  $\tau_{\lambda=0.856}$  and  $\alpha$  over ocean using a bi-  
35 modal aerosol size distribution (M. Wang et al., 2005). Further SeaWiFs aerosol products have been  
36 developed over both land and ocean using 6 and 8 visible channels respectively (e.g. von Hoyningen-Heune  
37 et al., 2003; Lee et al., 2004).

38 Despite the increased sophistication and realism of the aerosol retrieval algorithms, discrepancies exist  
39 between retrievals of  $\tau_{\text{aer}}$  even over ocean regions (e.g., Penner et al., 2002; Myhre et al., 2004a, 2005a;  
40 Jeong et al., 2005; Kinne et al., 2006). These discrepancies are due to different assumptions in the cloud  
41 clearing algorithms, aerosol models, different wavelengths, and viewing geometries used in the retrievals,  
42 different parameterisations of ocean surface reflectance, etc. Comparisons of these satellite aerosol retrievals  
43 with the surface AERONET observations provide an opportunity to objectively evaluate as well as improve  
44 the accuracy of these satellite retrievals. Myhre et al. (2005a) show that dedicated instruments using multi-  
45 channel and multi-view algorithms perform better when compared against AERONET than the simple  
46 algorithms that they have replaced, and Zhao et al. (2005) show that retrievals based on dynamic aerosol  
47 models perform better than those based on globally fixed aerosol models. While some systematic biases in  
48 specific satellite products exist (e.g., Jeong et al., 2005; Remer et al., 2005), these can be corrected for (e.g.,  
49 Bellouin et al., 2005; Kaufman et al., 2005b), which then enables an assessment of the direct radiative effect  
50 and the direct RF from an observational perspective, as detailed below.

#### 51 52 2.4.2.1.2 *Satellite retrievals of direct radiative effect, DRE.*

53 The solar direct radiative effect (DRE) is the sum of the direct effects due to anthropogenic and natural  
54 aerosol species while the direct RF only considers the anthropogenic components. Satellite estimates of the

1 global clear-sky DRE over oceans have advanced since the TAR owing to the development of dedicated  
2 aerosol instruments and algorithms, as summarised by Yu et al. (2006) (see Table 2.3). Table 2.3 suggests a  
3 reasonable agreement of the global-mean, diurnally averaged clear-sky DRE from various studies, with a  
4 mean of  $-5.4 \text{ W m}^{-2}$  and a standard deviation of  $0.9 \text{ W m}^{-2}$ . The clear-sky DRE is converted to an all-sky  
5 DRE by Loeb and Manalo-Smith (2005) who estimate an all sky DRE over oceans of  $-1.6$  to  $-2.0 \text{ W m}^{-2}$  but  
6 assume no aerosol contribution to the DRE from cloudy regions; such an assumption is not valid for  
7 optically thin clouds or if partially absorbing aerosols exist above the clouds (see Section 2.4.4.4).

8  
9 [INSERT TABLE 2.3 HERE]

10  
11 Furthermore, use of a combination of sensors on the same satellite offers the possibility of concurrently  
12 deriving  $\tau_{\text{aer}}$  and the DRE (e.g., Zhang and Christopher, 2003; Zhang et al., 2005), which enables estimation  
13 of the DRE efficiency, i.e., the DRE divided by  $\tau_{\text{aer}}$  ( $\text{W m}^{-2} \tau_{\text{aer}}^{-1}$ ). Because the DRE efficiency removes the  
14 dependence on the geographic distribution of  $\tau_{\text{aer}}$  it is a useful parameter for comparison of models against  
15 observations (e.g., Anderson et al., 2005b); however, the DRE efficiency thus derived is not a linear function  
16 of  $\tau_{\text{aer}}$  at high  $\tau_{\text{aer}}$  such as those associated with intense mineral dust, biomass burning or pollution events.

#### 17 18 2.4.2.1.3. *Satellite retrievals of direct radiative forcing*

19 Kaufman et al. (2005a) estimate the anthropogenic only component of the aerosol fine-mode fraction from  
20 the MODIS product to deduce a clear sky RF over ocean of  $-1.4 \text{ W m}^{-2}$ . Christopher et al. (2006) used a  
21 combination of the MODIS fine-mode fraction and CERES broadband TOA fluxes and estimated an  
22 identical value of  $-1.4 \pm 0.9 \text{ W m}^{-2}$ . Bellouin et al. (2005) use a combination of MODIS  $\tau_{\text{aer}}$  and fine-mode  
23 fraction together with data from AeroCom (see Section 2.4.3) to determine an all-sky RF of aerosols over  
24 both land and ocean of  $-0.8 \pm 0.2 \text{ W m}^{-2}$ , but this does not include the contribution to the RF and associated  
25 uncertainty from cloudy skies. Chung et al. (2005) perform a similar satellite/AERONET/model analysis, but  
26 include the contribution from cloudy areas to deduce an RF of  $-0.35 \text{ W m}^{-2}$  and  $-0.50 \text{ W m}^{-2}$  depending  
27 upon whether the anthropogenic fraction is determined from a model or from the MODIS fine-mode fraction  
28 and suggest an overall uncertainty of  $-0.1$  to  $-0.6 \text{ W m}^{-2}$ . Yu et al. (2006) used several measurements to  
29 estimate a direct RF of  $-0.5 \pm 0.33 \text{ W m}^{-2}$ . These estimates of the RF are compared to those obtained from  
30 modelling studies in Section 2.4.4.7.

#### 31 32 2.4.2.2 *Surface-Based Retrievals*

33  
34 A significant advancement since the TAR is the continued deployment and development of surface based  
35 remote sensing sun-photometer sites such as AERONET (Holben et al., 1998), and the establishment of  
36 networks of aerosol lidar systems such as the European Aerosol Research Lidar Network (EARLINET,  
37 Matthias et al., 2004), the Asian Dust Network (ADNET, Murayama et al., 2001), and the Micro-Pulse Lidar  
38 Network (MPLNET, Welton et al., 2001).

39  
40 The distribution of AERONET sites is also marked on Figure 2.11a. Currently there are approximately 150  
41 sites operating at any one time, many of which are permanent to enable determination of climatological and  
42 interannual column-averaged monthly and seasonal means. In addition to measurements of  $\tau_{\text{aer}}$  as a function  
43 of wavelength, new algorithms have been developed that measure sky radiance as a function of scattering  
44 angle (Nakajima et al., 1996; Dubovik and King, 2000). From these measurements, the column averaged size  
45 distribution and, if the  $\tau_{\text{aer}}$  is high enough ( $\tau_{\text{aer}} > 0.5$ ), the aerosol single scattering albedo,  $\omega_0$ , and refractive  
46 indices may be determined at particular wavelengths (Dubovik et al., 2000), allowing partitioning between  
47 scattering and absorption. Whilst these inversion products have not been comprehensively validated, a  
48 number of studies show encouraging agreement for both the derived size distribution and  $\omega_0$  when compared  
49 against in situ measurement by instrumented aircraft for different aerosol species (e.g., Dubovik et al., 2002;  
50 Haywood et al., 2003a; Reid et al., 2003; Osborne et al., 2004). A climatology of the aerosol DRE based on  
51 the AERONET aerosols has also been derived (Zhou et al., 2005).

52  
53 The MPLNET Lidar network currently consists of eleven lidars worldwide; nine are co-located with  
54 AERONET sites and provide complementary vertical distributions of aerosol backscatter and extinction.  
55 Additional temporary MPLNET sites have supported major aerosol field campaigns (e.g., Campbell et al.  
56 2003). EARLINET was a European-wide lidar network which currently has fifteen aerosol lidars making  
57 routine retrievals of vertical profiles of aerosol extinction (Mathias et al., 2004). ADNET is a network of

1 twelve lidars making routine measurements in Asia that have been used to assess the vertical profiles of  
2 Asian dust and pollution events (e.g., Husar et al., 2001; Murayama et al., 2001).

### 3 4 **2.4.3 Advances in Modelling the Aerosol Direct Effect**

5  
6 Since the TAR, more complete aerosol modules in a larger number of global atmospheric models now  
7 provide estimates of the direct RF. Several models have resolutions better than  $2^\circ \times 2^\circ$  degrees in the  
8 horizontal and more than 20 to 30 vertical levels; this represents a considerable enhancement over the  
9 models used in the TAR. Such models now include the most important anthropogenic and natural species.  
10 Tables 2.4, 2.5 and 2.6 summarise studies published since the TAR. Some of the more complex models  
11 account now explicitly for the dynamics of the aerosol size distribution throughout the aerosol atmospheric  
12 lifetime and also parameterise the internal/external mixing of the various aerosol components in a more  
13 physically realistic way than in the TAR (e.g. Adams and Seinfeld, 2002; Easter et al., 2004; Stier et al.,  
14 2005). Because the most important aerosol species are now included, a comparison of key model output  
15 parameters, such as the total  $\tau_{\text{aer}}$  is possible against satellite retrievals, and surface-based sun photometer and  
16 lidar observations (see Sections 2.4.2 and 2.4.4). Progress with respect to the modelling of the indirect  
17 effects due to aerosol-cloud interactions is detailed in Section 2.4.5 and Chapter 7 (Section 7.5). Several  
18 studies have explored the sensitivity of aerosol direct RF to current parameterisation uncertainties. These are  
19 assessed in the following sections.

20  
21 Major progress since the TAR has been made in the documentation of the diversity of current aerosol model  
22 simulations. Sixteen groups have participated in the Global Aerosol Model Intercomparison (AeroCom)  
23 initiative (Kinne et al., 2006). Extensive model outputs are available via a dedicated website (Schulz et al.,  
24 2004). Three model experiments (named *A*, *B*, *PRE*) were analysed: *Experiment A*: models simulate the  
25 years 1996, 1997, 2000 and 2001, or a five year mean encompassing these years. The model emissions and  
26 parameterisations are those determined by each research group, but the models are driven by observed  
27 meteorological fields to allow detailed comparisons with observations including those from MODIS, MISR  
28 and the AERONET sun photometer network.; *Experiment B*: models use prescribed AeroCom aerosol  
29 emissions for the year 2000; *Experiment PRE*: models use prescribed aerosol emissions for the year 1750  
30 (Dentener et al., 2006; Schulz et al. 2006). The model diagnostics included information on emission and  
31 deposition fluxes, vertical distribution and sizes, thus enabling a better understanding of the differences in  
32 lifetimes of the various aerosol components in the models.

33  
34 AeroCom results from Textor et al. (2006) are assessed in the remainder of this paragraph. The model  
35 comparison study finds a wide range in several of the diagnostic parameters; these, in turn, indicate which  
36 aerosol parameterisations are poorly constrained and/or understood. For example, coarse aerosol fractions  
37 are responsible for a large range in the natural aerosol emission fluxes (dust:  $\pm 49\%$ , sea salt:  $\pm 200\%$ , where  
38 uncertainty is  $1\sigma$  standard deviation of inter-model range), and consequently in the dry deposition fluxes.  
39 The complex dependence of the source strength on wind speed adds to the problem of computing natural  
40 aerosol emissions. Dust emissions for the same time period can vary by a factor of 2 or more depending on  
41 details of the dust parameterization (Luo et al., 2003; Timmreck and Schulz, 2004; Balkanski et al., 2004;  
42 Zender, 2004), and even depend on the reanalysis meteorological dataset used (Luo et al., 2003). With  
43 respect to anthropogenic and natural emissions of other aerosol components, modelling groups tended to  
44 make use of similar best guess information, e.g., recently revised emissions information available via the  
45 Global Emissions Inventory Activity (GEIA). The vertical aerosol distribution is shown to vary  
46 considerably, which is a consequence of important differences in removal and vertical mixing  
47 parameterisations. The inter-model range for the fraction of sulphate mass below 2.5 km to that of total  
48 sulphate is  $45\% \pm 23\%$ . Since humidification takes place mainly in the boundary layer, this source of inter-  
49 model variability increases the range of modelled direct RF. Additionally, differences in the parameterization  
50 of the wet deposition/vertical mixing process becomes more pronounced above 5 km altitude. Some models  
51 have a tendency to accumulate insoluble aerosol mass (dust and carbonaceous aerosols) at higher altitudes,  
52 while others have much more efficient wet removal schemes. Tropospheric residence times, defined here as  
53 the ratio of burden over sinks established for an equilibrated one-year simulation, vary by 20–30% for the  
54 fine-mode aerosol species. These variations are of interest, since they express the linearity of modelled  
55 emissions to aerosol burden and eventually to RF.

1 Considerable progress has been made in the systematic evaluation of global model results (see references in  
2 Tables 2.4–2.6). The simulated global  $\tau_{\text{aer}}$  at a wavelength of  $0.55\mu\text{m}$  in models ranges from 0.11 to 0.14.  
3 The values compare favourably to those obtained by remote sensing from the ground (AERONET ca. 0.135)  
4 and space (satellite composite ca. 0.15) (Kinne et al., 2003 and 2006), but significant differences exist in  
5 regional and temporal distributions. Modelled absorption optical thickness has been suggested to be  
6 underestimated by a factor of 2 to 4 when compared to observations (Sato et al., 2003) and DRE efficiencies  
7 have been shown to be lower in models both for the global average and regionally (Yu et al., 2006) (see  
8 Section 2.4.4.7). A merging of modelled and observed fields of aerosol parameters through assimilation  
9 methods of different degrees of complexity has also been performed since the TAR (e.g., Yu et al., 2003;  
10 Chung et al., 2005). Model results are constrained to obtain present day aerosol fields consistent with  
11 observations. Collins et al. (2001) showed that assimilation of satellite derived fields of  $\tau_{\text{aer}}$  can reduce the  
12 model bias down to 10% with respect to daily mean  $\tau_{\text{aer}}$  measured with a sun photometer at the INDOEX  
13 station Kaashidhoo. Liu et al. (2005) demonstrate similar efficient reduction of errors in  $\tau_{\text{aer}}$ . The magnitude  
14 of the global dust cycle has been suggested to range between 1500 and 2600  $\text{Tg yr}^{-1}$  by minimizing the bias  
15 between model and multiple dust observations (Cakmur et al. 2006). Bates et al. (2006) focused on three  
16 regions downwind of major urban/population centres and performed radiative transfer calculations  
17 constrained by intensive and extensive observational parameters to derive 24 hour average clear sky DRE of  
18  $-3.3 \pm 0.47$ ,  $-14 \pm 2.6$ ,  $-6.4 \pm 2.1 \text{ W m}^{-2}$  for the North Indian Ocean, the North West Pacific, and the North  
19 West Atlantic, respectively. By constraining aerosol models with these observations, the uncertainty  
20 associated with the DRE was reduced by approximately a factor of two.

#### 22 **2.4.4 Estimates of Aerosol Direct Radiative Forcing**

23  
24 Unless otherwise stated, this section discusses the TOA direct RF of different aerosol types as a global  
25 annual-mean quantity inclusive of the effects of clouds. Where possible, statistics from model results are  
26 used to assess the uncertainty in the RF. Recently published results and those grouped within AeroCom are  
27 assessed. Because AeroCom (B-PRE) results are based on prescribed emissions, the uncertainty in these  
28 results is lowered by having estimates of the uncertainties in the emissions. The quoted uncertainties  
29 therefore include the structural uncertainty (i.e., differences associated with the model formulation and  
30 structure) associated with the RF, but do not include the full range of parametric uncertainty (i.e., differences  
31 associated with the choice of key model parameters), as the model results are essentially best estimates  
32 constrained by observations of emissions, wet and dry deposition, size distributions, optical parameters,  
33 hygroscopicity, etc (Pan et al., 1997). The uncertainties are reported as the 5–95% confidence interval to  
34 allow the uncertainty in the RF of each species of aerosol to be quantitatively inter-compared.

##### 36 **2.4.4.1 Sulphate Aerosol**

37  
38 Atmospheric sulfate aerosol may be considered as consisting of sulphuric acid particles that are partly or  
39 totally neutralized by ammonia and are present as liquid droplets or partly crystallized. Sulphate is formed by  
40 aqueous phase reactions within cloud droplets, oxidation of  $\text{SO}_2$  via gaseous phase reactions with the  
41 hydroxyl radical, and by condensational growth onto pre-existing particles (e.g., Penner et al., 2001).  
42 Emission estimates are summarised by Haywood and Boucher (2000). The main source of sulphate aerosol is  
43 via sulphur dioxide emissions from fossil-fuel burning (about 72%), with a small contribution from biomass  
44 burning (about 2%) while natural sources are from dimethyl sulphide emissions by marine phytoplankton  
45 (about 19%) and by  $\text{SO}_2$  emissions from volcanoes (about 7%). Estimates of global  $\text{SO}_2$  emissions range  
46 from 66.8 to 92.4  $\text{TgS yr}^{-1}$  for anthropogenic emissions for the 1990s and from 91.7 to 125.5  $\text{TgS yr}^{-1}$  for  
47 total emissions. Emissions of sulphur dioxide from 25 countries in Europe have been reduced from  
48 approximately 18  $\text{TgS yr}^{-1}$  in 1980 to 4  $\text{TgS yr}^{-1}$  in 2002 (Vestreng et al, 2004). In the USA, the emissions  
49 have been reduced from about 10  $\text{TgS yr}^{-1}$  to 7  $\text{TgS yr}^{-1}$  (U. S. EPA, 2004: <http://www.epa.gov>) in the period  
50 1982–2001. However, over the same period  $\text{SO}_2$  emissions have been increasing significantly from Asia  
51 which is estimated to currently emit 17  $\text{TgS yr}^{-1}$  (Streets et al., 2003) and from developing countries in other  
52 regions (e.g., Lefohn et al., 1999; Van Aardenne et al., 2001, Boucher and Pham, 2002). The most recent  
53 study of Stern (2005) suggests a decrease in global anthropogenic emissions from approximately 73  $\text{TgS yr}^{-1}$   
54 to 54  $\text{TgS yr}^{-1}$  over the period 1980–2000, with northern hemisphere emission falling from 64  $\text{TgS yr}^{-1}$  to 43  
55  $\text{TgS yr}^{-1}$  and southern hemisphere emissions increasing from 9  $\text{TgS yr}^{-1}$  to 11  $\text{TgS yr}^{-1}$ . Smith et al. (2004)  
56 suggest a more modest decrease in global emissions, by some 10  $\text{TgS yr}^{-1}$  over the same period. The regional  
57 shift in the emissions of sulphur dioxide from U.S., Europe, Russia, Northern Atlantic Ocean and parts of

1 Africa to South-East Asia and the Indian and Pacific Ocean areas will lead to subsequent shifts in the pattern  
2 of the RF (e.g., Boucher and Pham, 2002, Smith et al., 2004; Pham et al., 2005). The recently used emission  
3 scenarios take into account effective injection heights and their regional and seasonal variability (e.g.,  
4 Dentener et al., 2006).

5  
6 The optical parameters of sulphate aerosol have been well documented (see Penner et al., 2001 and  
7 references therein). Sulphate is essentially an entirely scattering aerosol across the solar spectrum ( $\omega_0 = 1$ )  
8 but with a small degree of absorption in the near-infrared spectrum. Theoretical and experimental data are  
9 available on the relative humidity dependence of the specific extinction coefficient,  $f_{RH}$  (e.g., Tang et al.,  
10 1995). Measurement campaigns concentrating on industrial pollution such as TARFOX (Russell et al.,  
11 1999), ACE-2 (Raes et al., 2000), INDOEX (Ramanathan et al., 2001b), MINOS (2001 campaign), ACE-  
12 Asia (2001), APEX (2000-2003), NEAQS (2003), CLAMS (Smith et al., 2005) continue to show that  
13 sulphate contributes a significant fraction of the sub-micron aerosol mass, anthropogenic  $\tau_{aer}$  and RF (e.g.,  
14 Hegg et al., 1997; Russell and Heintzenberg, 2000; Ramanathan et al., 2001b; Magi et al., 2005; Quinn and  
15 Bates, 2005). However, sulphate is invariably internally and externally mixed to varying degrees with other  
16 compounds such as biomass burning aerosol (e.g., Formenti et al., 2003), fossil-fuel black carbon (e.g.,  
17 Russell and Heintzenberg, 2000), organic carbon (Novakov et al., 1997; Brock et al., 2004), mineral dust  
18 (e.g., Huebert et al., 2003), and nitrate aerosol (e.g., Schaap et al., 2004). This results in a composite aerosol  
19 state in terms of effective refractive indices, size distributions, physical state, morphology, hygroscopicity  
20 and optical properties.

21  
22 The TAR reported an RF due to sulphate aerosol of  $-0.40 \text{ W m}^{-2}$  with an uncertainty of a factor of two,  
23 based on global modelling studies that were available at that time. Results from model studies since the TAR  
24 are summarised in Table 2.4. For models A-L, the RF ranges from approximately  $-0.21 \text{ W m}^{-2}$  (Takemura et  
25 al., 2005) to  $-0.96 \text{ W m}^{-2}$  (Adams et al., 2001) with a mean of  $-0.46 \text{ W m}^{-2}$  and a standard deviation of  $0.20$   
26  $\text{W m}^{-2}$ . The range in the RF per unit  $\tau_{aer}$  is substantial due to differing representation of aerosol mixing state,  
27 optical properties, cloud, surface reflectance, hygroscopic growth, sub-gridscale effects, radiative transfer  
28 codes, etc. (Ramaswamy et al., 2001). Myhre et al. (2004b) performed several sensitivity studies finding that  
29 the uncertainty was particularly linked to the hygroscopic growth and that differences in the model relative  
30 humidity fields could cause differences of up to 60% in the RF. The RFs from the models M-U participating  
31 in the AeroCom project are slightly weaker than those obtained from the other studies with a mean of  
32 approximately  $-0.35 \text{ W m}^{-2}$  and a standard deviation of  $0.15 \text{ W m}^{-2}$ ; the standard deviation is reduced for the  
33 AeroCom models owing to constraints on aerosol emissions, based on updated emission inventories (see  
34 Table 2.4). Including the uncertainty in the emissions reported in Haywood and Boucher (2000) increases the  
35 standard deviation to  $0.2 \text{ W m}^{-2}$ . As sulphate aerosol is almost entirely scattering, the *surface forcing* will be  
36 similar or marginally stronger than the RF diagnosed at the top of the atmosphere. The uncertainty in the RF  
37 estimate relative to the mean value remains relatively large compared to the situation for LLGHGs.

38  
39 The mean and median of the sulfate direct RF from grouping all these studies together are identical at  $-0.41$   
40  $\text{W m}^{-2}$ . Disregarding the strongest and weakest direct RF estimates to approximate the 90% confidence  
41 interval leads to an estimate of  $-0.4 \pm 0.2 \text{ W m}^{-2}$ .

42  
43 [INSERT TABLE 2.4 HERE]

#### 44 45 2.4.4.2 *Organic Carbon Aerosol from Fossil Fuels*

46  
47 Organic aerosols are a complex mixture of chemical compounds containing carbon-carbon bonds produced  
48 from fossil-fuel and biofuel burning and natural biogenic emissions, either as primary aerosol particles or as  
49 secondary aerosol particles from condensation of low and semi-volatile organic gases. Hundreds of different  
50 atmospheric organic compounds have been detected in the atmosphere (e.g., Hamilton et al., 2004; Murphy,  
51 2005), which makes definitive modelling of the direct and indirect effects extremely challenging (McFiggans  
52 et al., 2006). Emissions of primary organic carbon from fossil fuel burning have been estimated to be 10 to  
53  $30 \text{ Tg C yr}^{-1}$  (Liousse et al., 1996; Cooke et al., 1999; Scholes and Andreae, 2000). More recently, Bond et  
54 al. (2004) provide a detailed analysis of primary organic carbon emissions from fossil-fuels, biofuels and  
55 open burning, and suggest that contained burning (approximately the sum of fossil-fuel and biofuel)  
56 emissions are in the range  $5\text{-}17 \text{ Tg C yr}^{-1}$ , with fossil-fuel contributing only  $2.4 \text{ Tg C yr}^{-1}$ . Ito and Penner  
57 (2005) estimated global fossil-fuel particulate organic matter (POM, which is the sum of the organic carbon

1 and the other associated chemical elements) emissions of around 2.2 Tg POM yr<sup>-1</sup>, and global biofuel  
2 emissions of around 7.5 Tg POM yr<sup>-1</sup>. Trends in emissions of fossil/bio fuel organic carbon have been  
3 estimated for the period 1870–2000 by Ito and Penner (2005) to have increased by a factor of three.  
4 Subsequent to emission, the hygroscopic, chemical, and optical properties of OC particles continue to change  
5 because of chemical processing by gas-phase oxidants such as O<sub>3</sub>, OH, and NO<sub>3</sub> (e.g. Kanakidou et al. 2005).  
6 Atmospheric concentrations of organic aerosol are frequently similar to those for industrial sulphate aerosol.  
7 Novakov et al. (1997) and Hegg et al. (1997) measured organic carbon in pollution off the East coast of the  
8 USA during the TARFOX campaign, and found organic carbon primarily from fossil fuel burning  
9 contributed up to 40% of the total submicron aerosol mass and was frequently the most significant  
10 contributor to  $\tau_{\text{aer}}$ . During INDOEX, which studied the industrial plume over the Indian Ocean, Ramanathan  
11 et al. (2001b) found organic carbon was the second largest contributor to  $\tau_{\text{aer}}$  after sulphate aerosol.  
12

13 Observational evidence suggests that some organic aerosol compounds from fossil fuels are relatively  
14 weakly absorbing but do absorb solar radiation at some ultra-violet and visible wavelengths (e.g., Bond et  
15 al., 1999; Jacobson, 1999; Bond, 2001) although organic aerosol from high temperature combustion such as  
16 fossil-fuel burning (Dubovik et al., 1998; Kirchstetter et al., 2004) appears less absorbing than from low  
17 temperature combustion such as open biomass burning. Observations suggest that a considerable fraction of  
18 organic carbon is soluble to some degree; whilst, at low relative humidity more water is often associated with  
19 the organic fraction than with inorganic material. At higher relative humidities, the hygroscopicity of organic  
20 carbon is considerably less than that of sulphate aerosol (Kotchenruther and Hobbs, 1998; Kotchenruther et  
21 al., 1999).  
22

23 Based on observations and fundamental chemical kinetic principles, attempts have been made to formulate  
24 organic carbon composition by functional group analysis in some main classes of organic chemical species  
25 (e.g., Decesari et al., 2000; Decesari et al., 2001; Maria et al., 2002; Ming and Russell, 2002), capturing  
26 some general characteristics in terms of refractive indices, hygroscopicity, and cloud activation properties.  
27 This facilitates improved parameterizations in global models (e.g., Fuzzi et al., 2001; Kanakidou et al., 2005;  
28 Ming et al., 2005a).  
29

30 Organic carbon aerosol from fossil fuel sources is invariably internally and externally mixed to some degree  
31 with other combustion products such as sulphate and black carbon (e.g., Novakov et al., 1997; Ramanathan  
32 et al., 2001b). Theoretically, coatings of essentially non-absorbing components such as organic carbon or  
33 sulphate on strongly absorbing core components such as black carbon can increase the absorption of the  
34 composite aerosol (e.g., Fuller et al., 1999; Jacobson, 2001a, Stier et al. 2006a), with results backed up by  
35 laboratory studies (e.g., Schnaiter et al., 2003). However coatings of organic carbon aerosol on hygroscopic  
36 aerosol such as sulphate may lead to suppression of the rate of water uptake during cloud activation (Xiong  
37 et al., 1998; Chuang, 2003).  
38

39 Current global models generally treat organic carbon using one or two tracers (e.g., water insoluble tracer,  
40 water soluble tracer) and highly parameterised schemes have been developed to represent the direct RF.  
41 Secondary organic carbon is highly simplified in the global models and in many cases treated as an  
42 additional source similar to primary organic carbon. Considerable uncertainties still exist in representing the  
43 refractive indices and the water of hydration associated with the particles because the aerosol properties will  
44 invariably differ depending on the combustion process, chemical processing in the atmosphere, mixing with  
45 the ambient aerosol etc (e.g., McFiggans et al., 2006).  
46

47 The TAR reported an RF of organic carbon aerosols from fossil fuel burning of  $-0.10 \text{ W m}^{-2}$  with a factor of  
48 three uncertainty. Many of the modelling studies that have been performed since the TAR have investigated  
49 the RF of organic carbon aerosols from both fossil-fuel and biomass burning aerosols, and the combined RF  
50 of both components. These studies are summarised in Table 2.5. The RF from total organic carbon (POM)  
51 from both biomass burning and fossil-fuel emissions from recently published models A-K and AeroCom  
52 models (L-T) is  $-0.24 \text{ W m}^{-2}$  with a standard deviation of  $0.08 \text{ W m}^{-2}$  and  $-0.16 \text{ W m}^{-2}$  with a standard  
53 deviation of  $0.10 \text{ W m}^{-2}$ , respectively. Where the RF due to organic carbon from fossil fuels is not explicitly  
54 accounted for in the studies, an approximate scaling based on the source apportionment of 0.25:0.75 is  
55 applied for fossil-fuel organic carbon: biomass burning organic carbon (Bond et al., 2004). The mean RF of  
56 the fossil-fuel component of organic carbon from those studies other than in AeroCom is  $-0.06 \text{ W m}^{-2}$ ,  
57 while those from AeroCom produce an RF of  $-0.03 \text{ W m}^{-2}$  with a range of  $-0.01 \text{ W m}^{-2}$  to  $-0.06 \text{ W m}^{-2}$  and

1 a standard deviation of around  $0.02 \text{ W m}^{-2}$ . Note that these RF estimates, to a large degree, only take into  
2 account primary emitted organic carbon. These studies all use optical properties for organic carbon that are  
3 either entirely scattering or only weakly absorbing and hence the *surface forcing* is only slightly stronger  
4 than that at the top of the atmosphere.  
5

6 The mean and median for the direct RF of fossil-fuel organic carbon from grouping all these studies together  
7 are identical at  $-0.05 \text{ W m}^{-2}$  with a standard deviation of  $0.03 \text{ W m}^{-2}$ . The standard deviation is multiplied by  
8 1.645 to approximate the 90% confidence interval<sup>10</sup>. This leads to a direct RF estimate of  $-0.05 \pm 0.05 \text{ W}$   
9  $\text{m}^{-2}$ .

10 [INSERT TABLE 2.5 HERE]  
11

#### 12 2.4.4.3 Black Carbon Aerosol from Fossil Fuels

13 Black carbon is a primary aerosol emitted directly at the source from incomplete combustion processes such  
14 as fossil-fuel and biomass burning and therefore much atmospheric BC is of anthropogenic origin. Global,  
15 current day, fossil-fuel emission estimates range from  $5.8 \text{ TgC yr}^{-1}$  to  $8.0 \text{ TgC yr}^{-1}$  (Haywood and Boucher,  
16 2000 and references therein). Bond et al. (2004) estimate the total current global emission of BC to be  
17 approximately  $8 \text{ TgC yr}^{-1}$  with contributions of  $4.6 \text{ TgC yr}^{-1}$  from fossil-fuel and biofuel combustion and  $3.3$   
18  $\text{TgC yr}^{-1}$  from open biomass burning and estimate an uncertainty of about a factor of two. Ito and Penner  
19 (2005) suggest fossil-fuel black carbon emissions for 2000 of around  $2.8 \text{ TgC yr}^{-1}$ . The trends in emission of  
20 fossil-fuel black carbon have been investigated in industrial areas by Novakov et al. (2003) and Ito and  
21 Penner (2005). Novakov et al. (2003) report significant decreases have been recorded in the UK, Germany,  
22 the former Soviet Union, and the USA over the period 1950–2000, while significant increases were reported  
23 in India and China. Globally, Novakov et al. (2003) suggest that emissions of fossil-fuel black carbon  
24 increased by a factor of three between 1950 and 1990 ( $2.2$  to  $6.7 \text{ TgC yr}^{-1}$ ) owing to the rapid expansion of  
25 the USA, European and Asian economies (e.g., Streets et al, 2001, 2003), and has since fallen to around  $5.6$   
26  $\text{TgC yr}^{-1}$  owing to further emission controls. Ito and Penner (2005) determined a similar trend in emissions  
27 over the period 1950–2000 of approximately a factor of three, but the absolute emissions are smaller than in  
28 Novakov et al. (2003) by approximately a factor of 1.7.  
29

30 Black carbon aerosol strongly absorbs solar radiation. Electron microscope images of BC particles show that  
31 BC particles are emitted as complex chain structures (e.g., Posfai et al., 2003), which tend to collapse as the  
32 particles age, thereby modifying the optical properties (e.g., Abel et al., 2003). The Indian Ocean Experiment  
33 (INDOEX, Ramanathan et al., 2001b and references therein) focussed on emissions of aerosol from the  
34 Indian sub-continent, and showed the importance of absorption by aerosol in the atmospheric column. Their  
35 observations showed that the local *surface forcing* ( $-23 \text{ W m}^{-2}$ ) was significantly stronger than the local RF  
36 at the top of the atmosphere ( $-7 \text{ W m}^{-2}$ ). Additionally, the presence of black carbon in the atmosphere above  
37 highly reflectant surfaces such as snow and ice, or clouds may cause a significant positive RF (Ramaswamy  
38 et al., 2001). The vertical profile is therefore important as black carbon aerosols or mixtures of aerosols  
39 containing a relatively large fraction of black carbon will exert a positive RF when located above clouds.  
40 Both microphysical (e.g., hydrophilic-to-hydrophobic nature of emissions into the atmosphere, aging of the  
41 aerosols, wet deposition) and meteorological aspects govern the horizontal and vertical distribution patterns  
42 of black carbon aerosols, and the residence time of these aerosols is thus sensitive to these factors (Cooke et  
43 al., 2002, Stier et al. 2006b).  
44

45 The TAR assessed the RF due to fossil-fuel black carbon as being  $+0.2 \text{ W m}^{-2}$  with an uncertainty of a factor  
46 of two. Those models since the TAR that explicitly model and separate out the RF due to black carbon from  
47 fossil fuels include those from Takemura et al. (2000), Reddy et al. (2005a), and Hansen et al. (2005) as  
48 summarised in Table 2.5. The results from a number of studies that continue to group the RF from fossil-fuel  
49 with that from biomass burning are also shown. Recently published results (A-K) and AeroCom studies (L-  
50 T) suggest a combined RF from both sources of  $+0.44 \pm 0.13 \text{ W m}^{-2}$  and  $+0.29 \pm 0.15 \text{ W m}^{-2}$  respectively.  
51 The stronger RF estimates from the models A-K appear to be primarily due to stronger sources and column  
52 loadings as the direct RF/column loading is similar at approximately  $1.2$  to  $1.3 \text{ W mg}^{-1}$  (Table 2.5).  
53 Carbonaceous aerosols emission inventories suggest approximately 34–38% of emissions come from  
54  
55

<sup>10</sup> 1.645 is the factor relating the standard deviation to the 90% confidence interval for a normal distribution

1 biomass burning sources and the remainder from fossil-fuel burning sources. Models which separate fossil  
2 fuel from biomass burning suggest a equal split in RF. This is applied to those estimates where the black  
3 carbon emissions are not explicitly separated into emission sources to provide an estimate of the RF due to  
4 fossil-fuel black carbon. For the AeroCom results, the fossil-fuel black carbon RF ranges from  $+0.08 \text{ W m}^{-2}$   
5 to  $+0.18 \text{ W m}^{-2}$  with a mean of  $+0.13 \text{ W m}^{-2}$  and a standard deviation of  $0.03 \text{ W m}^{-2}$ . For model results A-K,  
6 the RFs range from  $+0.15 \text{ W m}^{-2}$  to approximately  $+0.27 \text{ W m}^{-2}$ , with a mean of  $+0.25 \text{ W m}^{-2}$  and a standard  
7 deviation of  $0.11 \text{ W m}^{-2}$ .

8  
9 The mean and median of the direct RF for fossil-fuel black carbon from grouping all these studies together  
10 are  $+0.19 \text{ W m}^{-2}$  and  $+0.16 \text{ W m}^{-2}$  respectively with a standard deviation of nearly  $0.10 \text{ W m}^{-2}$ . The standard  
11 deviation is multiplied by 1.645 to approximate the 90% confidence interval and the best-estimate is rounded  
12 upwards slightly for simplicity, leading to a direct RF estimate of  $+0.20 \pm 0.15 \text{ W m}^{-2}$ . This estimate does  
13 not include the semi-direct effect or the BC impact on snow and ice surface albedo (see Sections 2.5.4 and  
14 2.8.5.6)

#### 15 2.4.4.4 Biomass Burning Aerosol

16 The TAR reported a contribution to the RF of roughly  $-0.4 \text{ W m}^{-2}$  from the scattering components (mainly  
17 organic carbon and inorganic compounds) and  $+0.2 \text{ W m}^{-2}$  from the absorbing components (BC) leading to  
18 an estimate of the RF of biomass burning aerosols of  $-0.20 \text{ W m}^{-2}$  with a factor of three uncertainty. Note  
19 that the estimates of the black carbon RF from Hansen and Sato (2001), Hansen et al. (2002), and Hansen  
20 and Nazarenko (2004) and Jacobson (2001a) include the RF component of BC from biomass burning  
21 aerosol. RF due to biomass burning (primarily organic carbon, black carbon, and inorganic compounds such  
22 as nitrate and sulphate) is grouped into a single RF, because biomass burning emissions are essentially  
23 uncontrolled. Emission inventories show more significant differences for biomass burning aerosols than for  
24 aerosols of from fossil fuel origin (Kasischke and Penner, 2004). Furthermore, the preindustrial levels of  
25 biomass burning aerosols are also difficult to quantify (Ito and Penner, 2005; Mouillot et al., 2006).

26  
27  
28 The Southern African Regional Science Initiative (SAFARI 2000: see Swap et al., 2002 and Swap et al.,  
29 2003) took place in 2000 and 2001. The main objectives of the aerosol research were to investigate  
30 pyrogenic and biogenic emissions of aerosol in southern Africa (Eatough et al., 2003; Formenti et al., 2003;  
31 Hély et al., 2003), validate the remote sensing retrievals (Haywood et al., 2003b; Ichoku et al., 2003), and to  
32 study the influence of aerosols on the radiation budget via the direct and indirect effects (e.g., Bergstrom et  
33 al., 2003; Keil and Haywood, 2003; Myhre et al., 2003; Ross et al., 2003). The physical and optical  
34 properties of fresh and aged biomass burning aerosol were characterised by making intensive observations of  
35 aerosol size distributions, optical properties, and DRE through in situ aircraft measurements (e.g., Abel et al.,  
36 2003; Formenti et al., 2003; Haywood et al., 2003b; Magi and Hobbs, 2003; Kirchstetter et al., 2004) and  
37 radiometric measurements (e.g., Bergstrom et al., 2003; Eck et al., 2003). The  $\omega_0$  at  $0.55 \mu\text{m}$  derived from  
38 near-source AERONET sites ranged from 0.85 to 0.89 (Eck et al., 2003), while  $\omega_0$  at  $0.55 \mu\text{m}$  for aged  
39 aerosol was less absorbing at approximately 0.91 (Haywood et al., 2003b). Abel et al. (2003) showed  
40 evidence that  $\omega_0$  at  $0.55 \mu\text{m}$  increased from approximately 0.85 to 0.90 over a time period of approximately  
41 two hours subsequent to emission, and attributed the result to the condensation of essentially non-absorbing  
42 organic gases onto existing aerosol particles. Fresh biomass burning aerosols produced by boreal forest fires  
43 appear to have weaker absorption than those from tropical fires, with  $\omega_0$  at  $0.55 \mu\text{m}$  greater than 0.9 (Wong  
44 and Li 2002). Boreal fires may not exert a significant direct RF because a large proportion of the fires are of  
45 natural origin and no significant change over the industrial era is expected. However, Westerling et al.  
46 (2006) showed that earlier spring and higher temperatures in USA have increased wild-fire activity and  
47 duration. The partially absorbing nature of biomass burning aerosol means it exerts an RF that is larger at the  
48 surface and in the atmospheric column than at the top of the atmosphere (see Figure 2.12).

49  
50 [INSERT FIGURE 2.12 HERE]

51  
52 Modelling efforts have used data from measurement campaigns to improve the representation of the physical  
53 and optical properties as well as the vertical profile of biomass burning aerosol (Myhre et al., 2003; Penner et  
54 al., 2003 and Section 2.4.5). These modifications have had important consequences on estimates of the RF  
55 due to biomass burning aerosols because the RF is significantly more positive when biomass burning aerosol  
56 overlies cloud than previously estimated (Keil and Haywood, 2003; Myhre et al., 2003; Abel et al., 2005).  
57 While the RF due to biomass burning aerosol in clear skies is certainly negative, the overall RF of biomass

burning aerosol may be positive. In addition to modelling studies, observations of this effect have been made with satellite instruments. 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  $100 \text{ W m}^{-2}$  and point 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  $\tau_{\text{aer}}$  and cloud effective radius in estimating the indirect radiative effect of aerosols.

Since the biomass burning aerosols can exert a significant positive RF when above clouds, the aerosol vertical profile is critical in assessing the magnitude and even the sign of the direct RF in cloudy areas. Textor et al. (2006) showed that there are significant differences in aerosol vertical profiles between global aerosol models. These differences are evident in the results from the recently published studies and AeroCom models in Table 2.5. The most negative RF of  $-0.05 \text{ W m}^{-2}$  is from the model of Koch (2001) and from the Myhre et al. (2003) AeroCom submission, while the most positive of  $+0.23 \text{ W m}^{-2}$  was derived in a simplistic manner in Schulz et al. (2006) from the ECHAM5-HAM AeroCom submission. Hence, even the sign of the RF due to biomass burning aerosols is in question.

The mean and median of the direct RF for biomass burning aerosol from grouping all these studies together are similar at  $+0.06 \text{ W m}^{-2}$  and  $+0.05 \text{ W m}^{-2}$  respectively with a standard deviation of  $0.08 \text{ W m}^{-2}$ . The standard deviation is multiplied by 1.645 to approximate the 90% confidence interval, leading to a direct RF estimate of  $+0.05 \pm 0.13 \text{ W m}^{-2}$ . This estimate of the direct RF is more positive than that of the TAR owing to improvements in the models in representing the absorption properties of the aerosol and the effects of biomass burning aerosol overlying clouds.

#### 2.4.4.5 Nitrate Aerosol

Atmospheric ammonium nitrate aerosol forms if sulphate aerosol is fully neutralised and there is excess ammonia. The direct RF due to nitrate aerosol is therefore sensitive to atmospheric concentrations of ammonia as well as  $\text{NO}_x$  emissions (Chapter 7, Section 7.4). In addition, the weakening of the RF of sulphate aerosol in many regions due to reduced emissions (Section 2.4.4.1) will be partially balanced by increases in the RF of nitrate aerosol (e.g., Liao and Seinfeld, 2005). The 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 (2001a) suggested relatively minor global mean RFs of  $-0.03 \text{ W m}^{-2}$  and  $-0.05 \text{ W m}^{-2}$ , respectively, while Adams et al. (2001) suggested a global mean RF as strong as  $-0.22 \text{ W m}^{-2}$ . 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  $-0.08 \text{ W m}^{-2}$  for global mean RF by nitrate. Further, Liao and Seinfeld (2005) estimated a global mean RF due to nitrate of  $-0.16 \text{ W m}^{-2}$ . In this study, heterogeneous chemistry reactions on particles were included; this strengthens the RF due to nitrate and accounts for 25% of its RF. Feng and Penner (2006) estimate a large, global, fine-mode nitrate burden of  $0.58 \text{ mg NO}_3 \text{ m}^{-2}$ , which would imply an equivalent of 20% of the mean anthropogenic sulphate burden. 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 1997; Martin et al. 2004 and references therein). 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 direct RF for nitrate is estimated to be  $-0.10 \text{ W m}^{-2}$  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, for example, of the nitrate  $\tau_{\text{aer}}$ . Thus a direct RF of  $-0.10 \pm 0.10 \text{ W m}^{-2}$  is tentatively adopted, but it is acknowledged that the number of studies performed is insufficient for accurate characterization of the magnitude and uncertainty of the RF.

#### 2.4.4.6 Mineral Dust Aerosol

1 Mineral dust from anthropogenic sources originates mainly from agricultural practices (harvesting,  
2 ploughing, over-grazing), changes in surface water (e.g., Caspian and Aral Sea, Owens Lake), and industrial  
3 practices (e.g., cement production, transport) (Prospero et al. 2002). The TAR reported that the RF due to  
4 anthropogenic mineral dust lies in the range of +0.4 to  $-0.6 \text{ W m}^{-2}$ , and did not assign a best estimate  
5 because of the difficulties in determining the anthropogenic contribution to the total dust loading and the  
6 uncertainties in the optical properties of dust and in evaluating the competing shortwave and longwave  
7 radiative effects. For the sign and magnitude of the mineral dust RF, the most important factor for the  
8 shortwave RF is the single scattering albedo whereas the longwave RF is dependent on the vertical profile of  
9 the dust.

10  
11 Tegen and Fung (1995) estimated the anthropogenic contribution to mineral dust to be 30–50% of the total  
12 dust burden in the atmosphere. Tegen et al. (2004) provided an updated, alternative estimate by comparing  
13 observations of visibility as a proxy for dust events from over 2000 surface stations with model results, and  
14 suggested that only 5–7% of mineral dust comes from anthropogenic agricultural sources. Yoshioka et al.  
15 (2005) suggested that a model simulation best reproduces the North African TOMS aerosol index  
16 observations when the cultivation source in the Sahel region contributes 0–15% to the total dust emissions in  
17 North Africa. A 35-year dust record established from Barbados surface dust and TOMS plus Meteosat  
18 satellite observations show the importance of climate control and Sahel drought for interannual and decadal  
19 dust variability, with no overall trend yet documented (Chiapello et al., 2005). As further detailed in Chapter  
20 7 (Section 7.3), climate change and carbon dioxide variations on various timescales can change vegetation  
21 cover in semi-arid regions. Such processes dominate over land-use changes as defined above, which would  
22 give rise to anthropogenic dust emissions (Mahowald and Luo, 2003; Moulin and Chiapello, 2004, Tegen et  
23 al., 2004). A best guess of 0–20% anthropogenic dust burden from these works is used here, but it is  
24 acknowledged that a very large uncertainty remains, because the methods used can not exclude either a  
25 reduction of 24% in present day dust nor a large anthropogenic contribution of up to 50% (Mahowald and  
26 Luo, 2003; Mahowald et al., 2004; Tegen et al., 2005). The RF efficiency of anthropogenic dust has not been  
27 well differentiated from that of natural dust and it is assumed that they are equal. The RF due to dust  
28 emission changes induced by circulation changes between 1750 and present day are difficult to quantify and  
29 not included here (see also Chapter 7, Section 7.5).

30  
31 In situ measurements of the optical properties of local Saharan dust (e.g., Haywood et al., 2003c; Tanré et  
32 al., 2003); transported Saharan mineral dust (e.g., Kaufman et al., 2001; Moulin et al., 2001; Coen et al.,  
33 2004) and Asian mineral dust (Huebert et al., 2003; Clarke et al., 2004; Mikami et al., 2006; Shi et al., 2005)  
34 reveal that dust is considerably less absorbing in the solar spectrum than suggested by previous dust models  
35 such as that of WMO (1986). These new, spectral, simultaneous remote and in situ observations suggest that  
36 the single scattering albedo ( $\omega_0$ ) of pure dust at a wavelength of  $0.67 \mu\text{m}$  is predominantly in the range 0.90–  
37 0.99, with a central global estimate of 0.96. This is in accordance with the bottom-up modelling of  $\omega_0$  based  
38 on the haematite content in desert dust sources (Claquin et al., 1999; Shi et al., 2005). Analyses of  $\omega_0$  from  
39 long-term AERONET sites influenced by Saharan dust suggest an average  $\omega_0$  of 0.95 at  $670\mu\text{m}$  (Dubovik et  
40 al., 2002), while unpolluted Asian dust during the Aeolian Dust Experiment on Climate (ADEC) has an  
41 average  $\omega_0$  of 0.93 at  $0.67\mu\text{m}$  (Mikami et al., 2006, and references therein). These high  $\omega_0$ -values suggest  
42 that a positive RF by dust in the solar region of the spectrum is unlikely. However, absorption by particles  
43 from source regions with variable mineralogical distributions is generally not represented by global models.

44  
45 Measurements of the DRE of mineral dust over ocean regions, where natural and anthropogenic  
46 contributions are indistinguishably mixed, suggest that the local DRE may be extremely strong; Haywood et  
47 al. (2003b) made aircraft-based measurements of the local instantaneous shortwave DRE of as strong as –  
48  $130 \text{ W m}^{-2}$  off the coast of West Africa. Hsu et al. (2000) used ERBE and TOMS data to determine a peak  
49 monthly mean shortwave DRE of around  $-45 \text{ W m}^{-2}$  for July 1985. Interferometer measurements from  
50 aircraft and the surface have now measured the spectral signature of mineral dust for a number of cases (e.g.,  
51 Highwood et al., 2003) indicating an absorption peak in the centre of the 8–13  $\mu\text{m}$  atmospheric window. Hsu  
52 et al. (2000) determined a longwave DRE over land areas of North Africa of up to  $+25 \text{ W m}^{-2}$  for July 1985;  
53 similar results were presented by Haywood et al. (2005) who determined a peak longwave DRE of up to  
54  $+50 \text{ W m}^{-2}$  at the top of the atmosphere for July 2003.

55  
56 Recent model simulations report the total anthropogenic and natural dust DRE, its components and the net  
57 effect as follows: (shortwave/longwave = net TOA) [ $\text{in W m}^{-2}$ ]: Liao et al. (2004):  $-0.21/+0.31 = +0.1$ ;

1 Reddy et al. (2005a):  $-0.28/+0.14 = -0.14$ ; Jacobson (2001a):  $-0.20/+0.07 = -0.13$ ; reference case and  
2 [range] of sensitivity experiments in Myhre and Stordal (2001a) (except case 6 and 7):  $-0.53[-1.4$  to  
3  $+0.2]/+0.13[+0.0$  to  $+0.8] = -0.4[-1.4$  to  $+1.0]$ ; and from AeroCom database: models GISS:  $-0.75/(+0.19) =$   
4  $(-0.56)$ ; UIO-CTM\*:  $-0.56/(+0.19) = (-0.37)$ ; LSCE\*:  $-0.6/+0.3 = -0.3$ ; UMI\*:  $-0.54/(+0.19) \Rightarrow (-0.35)$ .  
5 The (\*) star marked models use a single scattering albedo (approximately 0.96 at  $0.67\mu\text{m}$ ) that is more  
6 representative of recent measurements and show more negative shortwave effects. A mean longwave DRE of  
7  $0.19 \text{ W m}^{-2}$  is assumed for GISS, UMI and UIO-CTM. The scatter of dust DRE estimates reflects the fact  
8 that dust burden and  $\tau_{\text{aer}}$  vary by  $\pm 40\%$  and  $\pm 44\%$  respectively, computed as standard deviation from 16  
9 AeroCom A model simulations (Textor et al., 2006; Kinne et al., 2006). Dust emissions from different  
10 studies range between 1000 and  $2150 \text{ Tg yr}^{-1}$  (Zender, 2004). Finally, a major effect of dust may be in  
11 reducing the burden of anthropogenic species at sub-micron sizes and reducing their residence time (Bauer  
12 and Koch, 2005), see Section 2.4.5.7.

13  
14 The range of the reported dust net DRE ( $-0.56$  to  $+0.1 \text{ W m}^{-2}$ ), the revised anthropogenic contribution to  
15 dust DRE of 0–20% and the revised absorption properties of dust support a small negative value for the  
16 anthropogenic direct RF for dust of  $-0.1 \text{ W m}^{-2}$ . The 90% confidence level is estimated to be  $\pm 0.2 \text{ W m}^{-2}$ ,  
17 reflecting the uncertainty in total dust emissions and burdens and the range of possible anthropogenic dust  
18 fractions. At the limits of this uncertainty range, anthropogenic dust RF is as negative as  $-0.3 \text{ W m}^{-2}$  and as  
19 positive as  $+0.1 \text{ W m}^{-2}$ . This range includes all dust DRE reported above, assuming a maximum 20%  
20 anthropogenic dust fraction, except the most positive DRE from Myhre and Stordal (2001a).

#### 21 22 2.4.4.7 Direct RF for Combined Total Aerosol

23  
24 The TAR reported RF values associated with several aerosol components but did not provide an estimate of  
25 the overall aerosol RF. Improved and intensified in situ observations and remote sensing of aerosols suggest  
26 that the range of combined aerosol RF is now better constrained. For model results, extensive validation now  
27 exists for combined aerosol properties, representing the whole vertical column of the atmosphere, such as  
28  $\tau_{\text{aer}}$ . Using a combined estimate implicitly provides an alternative procedure to estimating the RF uncertainty.  
29 This approach may be more robust than propagating uncertainties from all individual aerosol components.  
30 Furthermore, a combined RF estimate accounts for non-linear processes due to aerosol dynamics and  
31 interactions between radiation field and aerosols. The role of non-linear processes of aerosol dynamics for  
32 RF has been recently studied in global aerosol models which account for the internally mixed nature of  
33 aerosol particles (Jacobson, 2001a; Kirkevåg and Iversen, 2002; Liao and Seinfeld, 2005; Stier et al., 2006b;  
34 Takemura et al., 2005). Mixing of aerosol particle populations influences the radiative properties of the  
35 combined aerosol, because mixing changes size, chemical composition, state, shape and this feed backs onto  
36 the aerosol removal and formation processes itself. Chung and Seinfeld (2002), in reviewing studies where  
37 black carbon is mixed either externally or internally with various other components, showed that black  
38 carbon exerts a stronger positive direct RF when mixed internally. Although the source-related processes for  
39 anthropogenic aerosols favour their submicron nature, natural aerosols enter the picture by providing a  
40 condensation surface for aerosol precursor gases. Heterogeneous reactions on sea salt and dust can reduce  
41 the sub-micron sulphate load by 28% (Liao et al., 2004) thereby reducing the direct and indirect RFs. Bauer  
42 and Koch (2005) estimate the sulphate RF to weaken from  $-0.25$  to  $-0.18 \text{ W m}^{-2}$  when dust is allowed to  
43 interact with the sulphur cycle. It would be useful to identify the RF contribution attributable to different  
44 source categories (Section 2.9.3 investigates this). However, few models have separated out the RF from  
45 specific emission source categories. Estimating the combined aerosol RF is a first step to quantify the  
46 anthropogenic perturbation to the aerosol and climate system caused by individual source categories.

47  
48 A central model-derived estimate for the aerosol direct RF is based here on a compilation of recent  
49 simulation results using multi-component global aerosol models (see Table 2.6). This is a robust method for  
50 several reasons: The complexity of multi-component aerosol simulations captures non-linear effects.  
51 Combining model results removes part of the errors in individual model formulations. As shown by Textor et  
52 al. (2006), the model-specific treatment of transport and removal processes is partly responsible for the  
53 correlated dispersion of the different aerosol components. A less dispersive model with smaller burdens  
54 necessarily has both less scattering and absorbing aerosols interacting with the radiation field. An error in  
55 accounting for cloud cover would affect the all-sky RF from all aerosol components. Such errors result in  
56 correlated RF efficiencies for major aerosol components within a given model. Directly combining aerosol  
57 RF results gives a more realistic aerosol RF uncertainty estimate. The AeroCom compilation suggests

1 significant differences in the modelled local and regional composition of the aerosol (see also Figure 2.12),  
2 but an overall reproduction of the total  $\tau_{\text{aer}}$  variability can be done (Kinne et al., 2006). The scatter in  
3 performance of the models suggests that currently no preference or weighting of individual model results can  
4 be used (Kinne et al., 2006). The aerosol RF taken together from several models is more robust than an  
5 analysis per component or by just one model. The mean estimate from Table 2.6 of the total aerosol direct  
6 RF is  $-0.2 \text{ W m}^{-2}$ , with a standard deviation of  $\pm 0.2 \text{ W m}^{-2}$ . This is a low-end estimate for both the aerosol  
7 RF and uncertainty because nitrate (estimated as  $-0.1 \text{ W m}^{-2}$ , see Section 2.4.4.5) and anthropogenic mineral  
8 dust (estimated as  $-0.1 \text{ W m}^{-2}$ , see Section 2.4.4.6) are missing in most of the model simulations. Adding  
9 their contribution yields an overall model-derived aerosol direct RF of  $-0.4 \text{ W m}^{-2}$  (90% confidence interval:  
10  $0 \text{ W m}^{-2}$  to  $-0.8 \text{ W m}^{-2}$ ).

11  
12 [INSERT TABLE 2.6]

13  
14 Three satellite-based measurement estimates of the aerosol direct RF have become available, which all  
15 suggest a more negative aerosol RF than the model studies (see Section 2.4.2.1.3). Bellouin et al. (2005)  
16 compute a TOA aerosol RF of  $-0.8 \pm 0.1 \text{ W m}^{-2}$ . Chung et al. (2005) based upon similarly extensive  
17 calculations estimate the value to be  $-0.35 \pm 0.25 \text{ W m}^{-2}$  and Yu et al. (2006)  $-0.5 \pm 0.33 \text{ W m}^{-2}$ . A central  
18 measurement based estimate would suggest a aerosol direct RF of  $-0.55 \text{ W m}^{-2}$ . Figure 2.13 shows the  
19 observational based aerosol direct RF estimates together with the published model estimates since the TAR.  
20 Also shown alongside are estimates from various modelling studies since the TAR.

21  
22 [INSERT FIGURE 2.13]

23  
24 The discrepancy between measurements and models is also apparent in oceanic clear-sky conditions where  
25 the measurement-based estimate of the combined aerosol DRE including natural aerosols is considered  
26 unbiased. In these areas models underestimate the negative aerosol DRE by 20–40% (Yu et al., 2006). The  
27 anthropogenic fraction of  $\tau_{\text{aer}}$  is similar between model and measurement based studies. Kaufman et al.  
28 (2005a) have used satellite observed fine-mode  $\tau_{\text{aer}}$  to estimate the anthropogenic  $\tau_{\text{aer}}$ . Correcting for fine-  
29 mode  $\tau_{\text{aer}}$  contributions from dust and sea salt, they find 21% of the total  $\tau_{\text{aer}}$  to be anthropogenic while Table  
30 2.6 suggests that 29% of  $\tau_{\text{aer}}$  is anthropogenic. Finally, cloud contamination of satellite products, aerosol  
31 absorption above clouds, not accounted for in some of the measurement-based estimates, and the complex  
32 assumptions on aerosol properties in both methods can contribute to the present discrepancy and increase  
33 uncertainty in aerosol RF.

34  
35 A large source of uncertainty in the aerosol RF estimates is associated with aerosol absorption. Sato et al.  
36 (2003) determined the absorption  $\tau_{\text{aer}}$  from AERONET measurements and suggested that aerosol absorption  
37 simulated by global aerosol models is underestimated by a factor of 2–4. Schuster et al. (2005) estimate the  
38 black carbon loading over continental scale regions. The results suggest that the model concentrations and  
39 absorption  $\tau_{\text{aer}}$  of black carbon from models are lower than those derived from AERONET. Some of this  
40 difference in concentrations could be explained by the assumption that all aerosol absorption is due to black  
41 carbon (Schuster et al., 2005), while a significant fraction may be due to absorption by organic aerosol and  
42 mineral dust (see Sections 2.4.4.2, and 2.4.4.6). Furthermore, Reddy et al. (2005a) show that comparison of  
43 the aerosol absorption  $\tau_{\text{aer}}$  from models against those from AERONET must be performed very carefully,  
44 reducing the discrepancy between their model and AERONET derived aerosol absorption  $\tau_{\text{aer}}$  from a factor  
45 of 4 to a factor of 1.2 by careful co-sampling of AERONET and model data. As mentioned above,  
46 uncertainty in the vertical position of absorbing aerosol relative to clouds can lead to large uncertainty in the  
47 top of the atmosphere aerosol RF.

48  
49 The partly absorbing nature of the aerosol is responsible for a heating of the lower tropospheric column and  
50 also results in the *surface forcing* being considerably more negative than top-of-the atmosphere RF, results  
51 that have been confirmed through several experimental and observational studies as discussed in earlier  
52 sections. Table 2.6 summarises the *surface forcing* obtained in the different models. Figure 2.12 depicts the  
53 regional distribution of several important parameters for assessing the regional impact of aerosol RF. The  
54 results are based on a mean model constructed from AeroCom simulation results B and PRE. Anthropogenic  
55  $\tau_{\text{aer}}$  (Figure 2.12a) is shown to have local maxima in industrialised regions and in biomass burning dominated  
56 areas. The difference between simulated and observed  $\tau_{\text{aer}}$  shows that regionally  $\tau_{\text{aer}}$  can be up to 0.1 (Figure

2.12b). Figure 2.12c suggests that there are regions off Southern Africa where the biomass burning aerosol above clouds leads to a local positive RF. Figure 2.12d shows the local variability as standard deviation from 9 models of the overall RF. The largest uncertainties of  $\pm 3 \text{ W m}^{-2}$  are to be found in East Asia and in the African biomass burning regions. Figure 2.12(e) reveals that an average of  $0.9 \text{ W m}^{-2}$  heating can be expected in the atmospheric column as a consequence of absorption by anthropogenic aerosols. Regionally, this can reach annually averaged values exceeding  $5 \text{ W m}^{-2}$ . These regional effects and the negative *surface forcing* in the shortwave (Figure 2.12f) is expected to exert an important effect on climate through alteration of the hydrological cycle.

An uncertainty estimate of the model derived aerosol direct RF can be based upon two alternative error analyses:

- 1) An error propagation analysis using the errors given in the sections 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 90% confidence level uncertainty of  $0.4 \text{ W m}^{-2}$ .
- 2) The standard deviation of the aerosol direct RF results in Table 2.6, multiplied by 1.645, suggests a 90% confidence level uncertainty of  $0.3 \text{ W m}^{-2}$  or  $0.4 \text{ W m}^{-2}$  when mineral dust and nitrate aerosol are accounted for.

Therefore, the results summarised in Table 2.6 and Figure 2.13, together with the estimates of nitrate and mineral dust RF combined with the measurement based estimates provide an estimate for the combined aerosol direct RF of  $-0.50 \pm 0.40 \text{ W m}^{-2}$ . The progress in both global modelling and measurements of the direct RF of aerosol leads to a *medium-low* level of scientific understanding (Table 2.11).

#### 2.4.5 Aerosol Influence on Clouds (Cloud Albedo Effect)

As pointed out in Section 2.4.1, aerosol particles affect the formation and properties of clouds. Only a subset of the aerosol population acts 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, Jacob et al., 2005). Several mechanisms are involved, as presented schematically in Figure 2.10. 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 mechanism is referred to in this report as the “cloud albedo effect”. The aerosol enhancements have also been hypothesised to lead to an increase in the lifetime of clouds (Albrecht, 1989); this mechanism is referred to in this report as the “cloud lifetime effect” and discussed in Chapter 7 (Section 7.5).

The interactions between aerosol particles (natural and anthropogenic in origin) and clouds are complex and can be non-linear (Ramaswamy et al., 2001). The size and chemical composition of the initial nuclei (e.g., anthropogenic sulphates, nitrates, dust, organic and black carbon) are important in the activation and early growth of the cloud droplets, particularly the water-soluble fraction and presence of compounds that affect surface tension (McFiggans et al., 2006 and references therein). Cloud optical properties are a function of wavelength. They depend on the characteristics of the droplet size distributions and ice crystal concentrations, and on the morphology of the various cloud types.

The interactions of increased concentrations of anthropogenic particles with shallow (stratocumulus and shallow cumulus) and deep convective clouds (with mixed phase) are discussed in this subsection. This section presents new observations and model estimates of the albedo effect. The associated RF in the context of liquid water clouds is assessed. In depth discussion of the induced changes that are not considered as RFs (e.g. semi-direct and cloud cover and lifetime effects, thermodynamic response, and changes in precipitation development) are presented in Chapter 7 (Section 7.5). The impacts of contrails and aviation-induced cirrus are discussed in Section 2.6 and the indirect impacts of aerosol on snow albedo are discussed in Section 2.5.4.

##### 2.4.5.1 Link Between Aerosol Particles and Cloud Microphysics

The local impact of anthropogenic aerosols has been known for a long time. For example, smoke from sugarcane and forest fires has been shown to reduce cloud droplet sizes in early case studies utilising in situ aircraft observations (Warner and Twomey, 1967; Eagan et al., 1974). On a regional scale, studies have

1 shown that heavy smoke from forest fires in the Amazon basin have led to increased cloud droplet number  
2 concentrations and to reduced cloud droplet sizes (Reid et al., 1999; Andreae et al., 2004, Mircea et al.,  
3 2005). The evidence concerning aerosol modification of clouds provided by the shiptrack observations  
4 reported in the TAR has been further confirmed, to a large extent qualitatively, by results from a number of  
5 studies using in situ aircraft and satellite data, covering continental cases and regional studies. Twohy et al.  
6 (2005) explored the relationship between aerosols and clouds in nine stratocumulus cases, indicating an  
7 inverse relationship between particle number and droplet size, but no correlation was found between albedo  
8 and particle concentration in the entire dataset. Feingold et al. (2003), Kim et al. (2003) and Penner et al.  
9 (2004) have presented evidence of an increase in the reflectance in continental stratocumulus cases, utilising  
10 remote sensing techniques at specific field sites. The estimates in Feingold et al. (2003) confirm that the  
11 relationship between aerosol and cloud droplet number concentrations is non-linear, e.g.,  $N_d \sim (N_a)^b$  where  $N_d$   
12 is the cloud drop number density and  $N_a$  is the aerosol number concentration. The parameter  $b$  in this  
13 relationship can vary widely, with values ranging from 0.06 to 0.48 (low values of  $b$  correspond to low  
14 hygroscopicity). This range highlights the sensitivity to aerosol characteristics (primarily size distribution),  
15 updraught velocity, and the usage of aerosol extinction as a proxy for CCN (Feingold, 2003). Disparity in the  
16 estimates of  $b$  (or equivalent) based on satellite studies (Nakajima et al. 2001; Breon et al. 2002) suggests  
17 that a quantitative estimate of the albedo effect from remote sensors is problematic (Rosenfeld and Feingold  
18 2003), particularly since measurements are not considered for similar liquid water paths.

19  
20 Many recent studies highlight the importance of aerosol particle composition in the activation process and  
21 droplet spectral evolution (indicated in the early laboratory work of Gunn and Phillips, 1957), but the picture  
22 that emerges is not complete. Airborne aerosol mass spectrometers provide firm evidence that ambient  
23 aerosols consist mostly of internal mixtures, e.g., biomass-burning components, organics and soot are mixed  
24 with other aerosol components (McFiggans et al., 2006). Mircea et al. (2005) showed the importance of the  
25 organic aerosol fraction in the activation of biomass burning aerosol particles. The presence of internal  
26 mixtures (e.g., sea-salt and organic compounds) can affect the uptake of water and the resulting optical  
27 properties compared to a pure sea-salt particle (Randles et al., 2004). Furthermore, the varying contents of  
28 water soluble and insoluble substances in internally mixed particles, the vast diversity of organics, and the  
29 resultant effects on cloud droplet sizes, makes the situation even more complex. Earlier observations of fog  
30 water (Facchini et al., 1999; Facchini et al., 2000) suggested that the presence of organic aerosols would  
31 reduce surface tension and lead to a significant increase in the cloud droplet number concentration (Nenes et  
32 al., 2002; Ming et al., 2005a; Rissler et al., 2004, McFiggans et al., 2006; Lohmann and Leck, 2005). On the  
33 other hand, Feingold and Chuang (2002) and Shantz et al. (2003) indicate that organic coating on CCN  
34 delayed activation, leading to a reduction in drop number and a broadening of the cloud droplet spectrum,  
35 which had not been previously considered. Ervens et al. (2005) address numerous composition effects in  
36 unison to show that the effect of composition on droplet number concentration is much less than suggested  
37 by studies that address individual composition effects, such as surface tension. The different relationships  
38 observed between cloud optical depth and liquid water path in clean and polluted stratocumulus clouds  
39 (Penner et al., 2004) have been explained by differences in sub-cloud aerosol particle distributions, while  
40 some contribution can be attributed to CCN composition (e.g., internally mixed insoluble dust, Asano et al.,  
41 2002). Nevertheless, the review by McFiggans et al. (2006) points to the remaining difficulties in explaining,  
42 quantitatively, the relationship between aerosol size and composition and the resulting droplet size  
43 distribution. Dusek et al. (2006) conclude that the ability of a particle to act as a CCN is largely controlled by  
44 size rather than composition.

45  
46 The complexity of the aerosol-cloud interactions and local atmospheric conditions where the clouds are  
47 developing are factors in the large variation evidenced for this phenomenon. Advances have been made in  
48 our understanding of the regional and/or global impact based on observational studies, particularly for low-  
49 level stratiform clouds that constitute a simpler cloud system to study than many of the other cloud types.  
50 Column aerosol number concentration and column cloud droplet concentration over the oceans from the  
51 Advanced Very High Resolution Radiometer, AVHRR, (Nakajima et al., 2001) indicate a positive  
52 correlation, and an increase in shortwave reflectance of low-level, warm clouds with increasing cloud optical  
53 thickness, while liquid water path (LWP) remained unmodified. While these results are only applicable over  
54 the oceans and are based on data for only four months, the positive correlation between an increase in cloud  
55 reflectance and an enhanced ambient aerosol concentration has been confirmed by other studies (Brennguier  
56 et al., 2000a; Brennguier et al., 2000b; Rosenfeld et al., 2002). However, other studies highlight the sensitivity  
57 to liquid water path (LWP), linking high pollution entrained into clouds to a decrease in LWP and a

1 reduction in the observed cloud reflectance (Jiang et al., 2002; Brenguier et al., 2003, Twohy et al., 2005).  
2 Still others (Han et al., 2002, using AVHRR observations) have reported an absence of LWP changes in  
3 response to increases of the column-averaged droplet number concentration ( $N_c$ ); this occurred for one-third  
4 of the cloud cases studied whose optical depths ranged between 1 and 15. Results of large eddy simulations  
5 of stratocumulus (Jiang et al. 2002; Ackerman et al. 2004; Lu and Seinfeld, 2005) and cumulus clouds (Xue  
6 and Feingold 2006; Jiang and Feingold, 2006) seem to confirm the lack of increase in LWP due to increases  
7 in aerosols; they point to a dependence on precipitation rate and relative humidity above the clouds  
8 (Ackerman et al, 2004). The studies above highlight the difficulty of devising observational studies that can  
9 isolate the albedo effect from other effects (e.g., meteorological variability, cloud dynamics) that influence  
10 LWP and therefore cloud RF.

11  
12 Results from the POLDER satellite instrument, which retrieve both submicron aerosol loading and cloud  
13 droplet size, suggest much larger cloud effective radii in remote oceanic regions than in the highly polluted  
14 continental source areas and downwind adjacent oceanic areas, namely from a maximum of 14 down to 6  
15  $\mu\text{m}$  (Bréon et al., 2002). This confirms earlier studies of hemispheric differences using AVHRR. Further, the  
16 POLDER- and AVHRR-derived correlations between aerosol and cloud parameters are consistent with an  
17 aerosol indirect effect (Sekiguchi et al. (2003). These results suggest that the impact of aerosols on cloud  
18 microphysics is global. Note that the satellite measurements of aerosol loading and cloud droplet size are not  
19 coincident, and an aerosol index is not determined in the presence of clouds. Further, there is a lack of  
20 simultaneous measurements of LWP which makes the assessment of the cloud albedo RF difficult,  
21

22 The albedo effect is also estimated from studies that combined satellite retrievals with a chemical transport  
23 model e.g., in the case of two pollution episodes over the mid-latitude Atlantic Ocean. Results indicate a  
24 brightening of clouds over a timescale of a few days in instances when liquid water path did not undergo any  
25 significant changes (Hashvardhan et al., 2002; Schwartz et al., 2002; Krüger and Graßl, 2002). There have  
26 been fewer studies on aerosol-cloud relationships under more complex meteorological conditions e.g.,  
27 simultaneous presence of different cloud types.  
28

29 The presence of insoluble particles within ice crystals constituting clouds formed at cold temperatures has a  
30 significant influence on the radiation transfer. The inclusions of scattering and absorbing particles within  
31 large ice crystals (Macke et al., 1996), suggest a significant effect. Hence, when soot particles are embedded,  
32 there is an increase in the asymmetry parameter and thus forward scattering. In contrast, inclusions of  
33 ammonium sulphate or air bubbles lead to a decrease in the asymmetry parameter of ice clouds Given the  
34 recent observations of partially insoluble nuclei in ice crystals (Cziczo et al., 2004) and the presence of small  
35 crystal populations, there is a need to further develop the solution for radiative transfer through such systems.  
36

#### 37 2.4.5.2 *Estimates of the RF from Models*

38  
39 General circulation models (GCMs) constitute an important and useful tool to estimate the global mean RF  
40 associated with the cloud albedo effect of anthropogenic aerosols. The model estimates of the changes in  
41 cloud reflectance are based on forward calculations, considering emissions of anthropogenic primary  
42 particles and secondary particle production from anthropogenic gases. Since the TAR, the cloud albedo  
43 effect has been estimated in a more systematic and rigorous way (allowing, for example, for the relaxation of  
44 the fixed LWC criterion), and more modelling results are now available. Most climate models use  
45 parameterisations to relate the cloud droplet number concentration to aerosol concentration; these vary in  
46 complexity from simple empirical fits to more physically-based relationships. Some models are run under an  
47 increasing greenhouse gas concentration scenario and include estimates of present day aerosol loadings  
48 (including primary and secondary aerosol production from anthropogenic sources). These global modelling  
49 studies (Table 2.7) have a limitation arising due to the underlying uncertainties in aerosol emissions (e.g.,  
50 emission rates of primary particles and of secondary particle precursors). Another limitation is the inability  
51 to perform a meaningful comparison between the various model results owing to differing formulations of  
52 relationships between aerosol particle concentrations and cloud droplet or ice crystal populations; this, in  
53 turn, yields differences in the impact of microphysical changes on the optical properties of clouds. Further,  
54 even when the relationships used in different models are similar, there are noticeable differences in the  
55 spatial distributions of the simulated low-level clouds. Individual models' physics have undergone  
56 considerable evolution, and it is difficult to clearly identify all the changes in the models as they have  
57 evolved. While GCMs have other well-known limitations such as: coarse spatial resolution, accurate

1 representation of convection and hence updraft velocities leading to aerosol activation and cloud formation  
2 processes, and microphysical parameterizations, they nevertheless remain an essential tool for quantifying  
3 the global cloud albedo effect. In Table 2.7, differences in the treatment of the aerosol mixtures (internal vs.  
4 external, with the latter being the more frequently employed method) are noted. Case studies of droplet  
5 activation indicate a clear sensitivity to the aerosol composition (McFiggans et al, 2006); additionally,  
6 radiative transfer is sensitive to the aerosol composition and the insoluble fraction present in the cloud  
7 droplets.

8  
9 [INSERT TABLE 2.7 HERE]

10  
11 All models estimate a negative global-mean RF associated with the cloud albedo effect, with the range of  
12 model results varying widely, from  $-0.22$  to  $-1.85$   $\text{W m}^{-2}$ . There are considerable differences in the  
13 treatment of aerosol, cloud processes and aerosol-cloud interactions processes in these models. Several  
14 models include an interactive sulphur cycle and anthropogenic aerosol particles composed of sulphate, as  
15 well as naturally produced sea-salt, dust and continuously outgassing volcanic sulphate aerosols. Lohmann et  
16 al. (2000) and Chuang et al. (2002) include internally mixed sulphate, black and organic carbon, sea salt and  
17 dust aerosols, resulting in the most negative estimate of the cloud albedo indirect effect. Takemura et al.  
18 (2005) use a global aerosol transport-radiation model coupled to a general circulation model, to estimate the  
19 direct and indirect effects of aerosols and their associated RF. The model includes a microphysical  
20 parameterisation to diagnose the cloud droplet number concentration using Köhler theory, which depends on  
21 the aerosol particle number concentration, updraft velocity, size distributions, and chemical properties of  
22 each aerosol species. The results indicate a global decrease in cloud droplet effective radius caused by  
23 anthropogenic aerosols, with the global mean RF calculated to be  $-0.52$   $\text{W m}^{-2}$ ; the land and oceanic  
24 contributions are  $-1.14$  and  $-0.28$   $\text{W m}^{-2}$ , respectively. Other modelling results also indicate that the mean  
25 RF due to the cloud albedo effect is on average somewhat larger over land than over oceans; over oceans  
26 there is a more consistent response from the different models, resulting in a smaller inter-model variability  
27 (Lohmann and Feichter, 2005).

28  
29 Chen and Penner (2005), by systematically varying parameters, obtain a less negative RF when the in-cloud  
30 updraft velocity is made to depend on the turbulent kinetic energy ( $W\_TKE$ ). Incorporating other cloud  
31 nucleation schemes e.g., changing from Abdul-Razzak and Ghan (2002) to the Chuang et al. (1997)  
32 parameterization results in no RF change, while changing to the Nenes and Seinfeld (2003) parameterization  
33 makes the RF more negative. Rotstayn and Liu (2003) found a 12 to 35 % decrease in the RF when the size  
34 dispersion effect was included in the case of sulphate particles. Chen and Penner (2005) further explore the  
35 range of parameters used in Rotstayn and Liu (2003) and found the RF to be generally less negative than in  
36 the standard integration.

37  
38 A model intercomparison study (Penner et al., 2006a) has examined the differences in cloud albedo effect  
39 between models through a series of controlled experiments which allows examination of the uncertainties.  
40 This study presents results from three models, which were run with prescribed aerosol mass-number  
41 concentration (from Boucher and Lohmann, 1995), aerosol field (from Chen and Penner, 2005) and  
42 precipitation efficiency (from Sundqvist, 1978). The cloud albedo RFs in the three models do not vary  
43 widely:  $-0.65$ ,  $-0.68$  and  $-0.74$   $\text{W m}^{-2}$ , respectively. Nevertheless, changes in the autoconversion scheme led  
44 to a differing response of the liquid water path between the models, and this is identified as an uncertainty.

45  
46 A closer inspection of the treatment of aerosol species in the models leads to a broad separation of the results  
47 into two groups: models with only a few aerosol species versus those that include a more complex mixture of  
48 aerosols of different composition. Thus, in Figure 2.14, RF results are grouped according to the type of  
49 aerosol species included in the simulations. In the top panel of Figure 2.14, where estimates from models that  
50 mainly include anthropogenic sulphate are shown, there is an indication that the results are converging, even  
51 though the range of models comes from studies published between 2001 and 2006. These studies show much  
52 less scatter than in the TAR, with a mean and standard deviation of  $-1.37 \pm 0.14$   $\text{W m}^{-2}$ . In contrast, in the  
53 bottom panel of Figure 2.14, that shows the studies which include more species, a much larger variability is  
54 found. These latter models (see Table 2.7) include “state of the art” parameterisations of droplet activation  
55 for a variety of aerosols, and include both internal and external mixtures.

56  
57 [INSERT FIGURE 2.14 HERE]

1  
2 Some studies have commented on inconsistencies between some of the earlier estimates of the cloud albedo  
3 RF from forward and inverse calculations (Anderson et al., 2003). Notwithstanding the fact that these two  
4 streams of calculations rely on very different formulations, the results here appear to be within range of the  
5 estimates from inverse calculations.  
6

#### 7 *2.4.5.3 Estimates of the RF from Observations and Constrained Models*

8

9 It is difficult to obtain a best estimate of the cloud albedo RF from pre-industrial times to present-day based  
10 solely on observations. The satellite record is not long enough, and other long-term records do not provide  
11 the pre-industrial aerosol and cloud microphysical properties needed for such an assessment. Some studies  
12 have attempted to estimate the RF by incorporating empirical relationships derived from satellite  
13 observations. This approach is valid as long as the observations are robust, but problems still remain,  
14 particularly with the use of the aerosol optical depth as proxy for CCN (Feingold et al., 2003), droplet size  
15 and cloud optical depth from broken clouds (Marshak et al., 2006), and relative humidity effects (Kapustin et  
16 al., 2006) to discriminate between hydrated aerosols and cloud. RF estimates constrained by satellite  
17 observations need to be considered with these caveats in mind.  
18

19 By assuming a bimodal log-normal size distribution, Nakajima et al. (2001) determine the Angstrom  
20 exponent from AVHRR data over the oceans (for a period of four months), together with cloud properties,  
21 optical thickness and effective radii. The non-linear relationship between aerosol number concentration and  
22 cloud droplet concentration ( $N_d \sim (N_a)^b$ ) obtained is consistent with Twomey's hypothesis; however, the  
23 parameter  $b$  is smaller than previous estimates (0.5 versus 0.7–0.8; Kaufman et al., 1991), but larger than the  
24 0.26 value obtained by Martin et al. (1994). Using this relationship, they provide an estimate of the cloud  
25 albedo RF in the range between  $-0.7$  and  $-1.7 \text{ W m}^{-2}$ , with a global average of  $-1.3 \text{ W m}^{-2}$ . Lohmann and  
26 Lesins (2002) used POLDER data to estimate aerosol index and cloud droplet radius; they then scaled the  
27 results of the simulations with the ECHAM4 model. The results show that changes in  $N_a$  lead to larger  
28 changes of  $N_d$  in the model than in observations, particularly over land, leading to an overestimate of the  
29 cloud albedo effect. The scaled values using the constraint from POLDER yield a global cloud albedo RF of  
30  $-0.85 \text{ W m}^{-2}$ , an almost 40% reduction from their previous estimate. Sekiguchi et al. (2003) presents results  
31 from the analysis of AVHRR data over the oceans, and of POLDER data over land and ocean. Assuming that  
32 the aerosol column number concentration increased by 30% from the pre-industrial era, they estimate the  
33 effect due to the aerosol influence on clouds as the difference between the forcing under present and pre-  
34 industrial conditions. They estimate a global effect due to the total aerosol influence on clouds (sum of cloud  
35 albedo and lifetime effects) to be between  $-0.6$  and  $-1.2 \text{ W m}^{-2}$ , somewhat lower than the Nakajima et al.  
36 (2001) ocean estimate. When the assumption is made that the liquid water content is constant, the cloud  
37 albedo RF estimated from AVHRR data is  $-0.64 \pm 0.16 \text{ W m}^{-2}$ ; the estimate using POLDER data is  $-0.37 \pm$   
38  $0.09 \text{ W m}^{-2}$ . The results from these two studies are very sensitive to the magnitude of the increase in the  
39 aerosol concentration from pre-industrial to current conditions, and the spatial distributions.  
40

41 Quaas and Boucher (2005) used the POLDER and MODIS data to evaluate the relationship between cloud  
42 properties and aerosol concentrations on a global scale in order to incorporate it in a GCM. They derive  
43 relationships corresponding to marine stratiform clouds and convective clouds over land which show a  
44 decreasing effective radius as the aerosol optical depth increases. These retrievals involve a variety of  
45 assumptions that introduce uncertainties in the relationships, in particular the fact that the retrievals for  
46 aerosol and cloud properties are not coincident and the assumption that the aerosol optical depth can be  
47 linked to the sub-cloud aerosol concentration. When these empirical parameterisations are included in a  
48 climate model, the simulated RF due to the cloud albedo effect is reduced by 50% from their baseline  
49 simulation. Quaas et al. (2005) also utilise satellite data to establish a relationship between cloud droplet  
50 number concentration and fine mode aerosol optical depth, minimising the dependence on cloud liquid water  
51 content but including an adiabatic assumption that may not be realistic in many cases. This relationship is  
52 implemented in the ECHAM4 and LMDZ climate models and the results indicate that the original  
53 parameterisations used in both models overestimated the magnitude of the cloud albedo effect. Even though  
54 both models show a consistent weakening of the RF, it should be noted that the original estimates of their  
55 respective RFs are very different (by almost a factor of two); the amount of the reduction was 37 % in  
56 LMDZ and 81% in ECHAM4. Note that the two models have highly different spatial distributions of low  
57 clouds, simulated aerosol concentrations and anthropogenic fractions.

1  
2 When only sulphate aerosols are considered, Dufresne et al. (2005) obtain a weaker cloud albedo RF. Their  
3 model uses a relationship between aerosol mass concentration and cloud droplet number concentration,  
4 modified from that originally proposed by Boucher and Lohmann (1995) and adjusted to POLDER data.  
5 Their simulations give a factor of two weaker RF compared to the previous parameterisation, but it is noted  
6 that the results are highly sensitive to the distribution of clouds over land.  
7

#### 8 *2.4.5.4 Uncertainties in Satellite Estimates*

9

10 The improvements in the retrievals and satellite instrumentation have provided valuable data to begin  
11 observational-motivated assessments of the impact of the effect of aerosols on cloud properties, even though  
12 satellite measurements cannot distinguish unambiguously natural from anthropogenic aerosols. Nevertheless,  
13 an obvious advantage of the satellite data is their global coverage, and such extensive coverage can be  
14 analysed to determine the relationships between aerosol and cloud properties at a number of locations around  
15 the globe. Using these data some studies (Sekiguchi et al., 2003; Quaas et al., 2004) indicate that the  
16 magnitude of the RF is resolution-dependent, since the representation of convection and clouds in the GCMs  
17 and the simulation of updraught velocity that affects activation themselves are resolution-dependent. The  
18 rather low spatial and temporal resolution of some of the satellite datasets can introduce biases by failing to  
19 distinguish aerosol species with different properties. This, together with the absence of coincident liquid  
20 water path measurements in several instances, handicaps the inferences from such studies, and hinders an  
21 accurate analysis and estimate of the RF. Furthermore, the ability to separate meteorological from chemical  
22 influences in satellite observations depends on the understanding of how clouds respond to meteorological  
23 conditions.  
24

25 Retrievals involve a variety of assumptions that introduce uncertainties in the relationships. As mentioned  
26 above, the retrievals for aerosol and cloud properties are not coincident and the assumption is made that the  
27 aerosol optical depth can be linked to the aerosol concentration below the cloud. POLDER may  
28 underestimate the mean cloud top droplet radius due to uncertainties in the sampling of clouds (Rosenfeld  
29 and Feingold, 2003). The retrieval of the aerosol index over land may be less reliable and lead to an  
30 underestimate of the cloud albedo effect over land. There is an indication of a systematic bias between  
31 MODIS-derived cloud droplet radius and that derived from POLDER (Breon and Doutriaux-Boucher, 2005),  
32 as well as differences in the aerosol optical depth retrieved from those instruments (Myhre et al., 2004a) that  
33 need to be resolved.  
34

#### 35 *2.4.5.5 Uncertainties Due to Model Biases*

36

37 One of the large sources of uncertainties is the poor knowledge of the amount and distribution of  
38 anthropogenic aerosols used in the model simulations, particularly for pre-industrial conditions. Some  
39 studies show a large sensitivity in the RF to the ratio of pre-industrial to present day aerosol number  
40 concentrations.  
41

42 All climate models discussed above include sulphate particles; some models produce them from gaseous  
43 precursors over oceans, where ambient concentrations are low; some models only condense mass onto pre-  
44 existing particles over the continents. Some other climate models also include sea-salt and dust particles  
45 produced naturally, typically relating particle production in terms of wind speed. Some models include  
46 anthropogenic nitrate, black carbon and organic compounds, which in turn affect activation. Models also  
47 have weaknesses in representing convection processes, aerosol distributions, and simulating updraught  
48 velocities and convection-cloud interactions. Even without considering the existing biases in the model-  
49 generated clouds, differences in the aerosol chemical composition and the subsequent treatment of activation  
50 lead to uncertainties that are difficult to quantify and assess. The presence of organic carbon owing to its  
51 distinct hygroscopic and absorption properties can be particularly important for the cloud albedo effect in the  
52 tropics (Ming et al., 2006b).  
53

54 Modelling the cloud albedo effect from first principles has proven difficult because the representation of  
55 aerosol-cloud and convection-cloud interactions in climate models are still crude (Lohmann and Feichter,  
56 2005). Clouds often do not cover a complete grid box and are inhomogeneous in terms of droplet  
57 concentration, effective radii and LWP, which introduces added complications in the microphysical and

1 radiative transfer calculations. Model intercomparisons (e.g., Lohmann et al., 2001; Menon et al., 2003)  
2 suggest that the predicted cloud distributions vary significantly between models, particularly their horizontal  
3 and vertical extents; also, the vertical resolution and parameterisation of convective and stratiform clouds are  
4 quite different between models (Chen and Penner, 2005). Even high resolution models have difficulty in  
5 accurately estimating the amount of cloud liquid and ice water content in a grid box.  
6

7 It has proven difficult to compare directly the results from the different models, as uncertainties are not well  
8 identified and quantified. All models could be suffering from similar biases, and modelling studies do not  
9 often quote the statistical significance of the RF estimates that are presented. Ming et al. (2005b)  
10 demonstrate that it is only in the midlatitude northern hemisphere that their model yields a RF result at the  
11 95% confidence level when compared to the unforced model variability. There are also large differences in  
12 the way that the different models treat the appearance and evolution of aerosol particles and the subsequent  
13 cloud droplet formation. Differences in the horizontal and vertical resolution introduce uncertainties in their  
14 ability to accurately represent the shallow warm cloud layers over the oceans that are most susceptible to the  
15 changes due to anthropogenic aerosol particles. A more fundamental problem is that GCMs do not resolve  
16 the small scales (order of 100s of metres) at which aerosol-cloud interactions occur. Chemical composition  
17 and size distribution spectrum are also likely insufficiently understood on a microphysical level, although  
18 some modelling studies suggest that the albedo effect is more sensitive to the size than to aerosol  
19 composition (Feingold, 2003; Ervens et al., 2005; Dusek et al., 2006). Observations indicate that aerosol  
20 particles in nature tend to be composed of several compounds and can be internally or externally mixed. The  
21 actual conditions are difficult to simulate and possibly lead to differences amongst climate models. The  
22 calculation of the cloud albedo effect is sensitive to the details of particle chemical composition (activation)  
23 and state of the mixture (external vs. internal). The relationship between ambient aerosol particle  
24 concentrations and resulting cloud droplet size distribution is important during the activation process; this is  
25 a critical parameterisation element in the climate models. It is treated in different ways in different models,  
26 ranging from simple empirical functions (Menon et al., 2002b), to more complex physical parameterisations  
27 that also tend to be more costly computationally (Abdul-Razzak and Ghan, 2002; Nenes and Seinfeld, 2003;  
28 Ming et al., 2006a). Finally, comparisons with observations have not yet risen to the same degree of  
29 verification as, for example for the direct RF estimates; this is not merely due to model limitations, since the  
30 observational basis also has not yet reached a sound footing.  
31

32 Further uncertainties may arise be due to changes in the droplet spectral shape, typically considered invariant  
33 in climate models under clean and polluted conditions, but which can be substantially different in typical  
34 atmospheric conditions (e.g., Feingold et al, 1997; Ackerman et al, 2000b; Erlick et al., 2001; Liu and Daum,  
35 2002). Liu and Daum (2002) estimated that a 15% increase in the width of the size distribution can lead to a  
36 reduction of between 10 and 80% in the estimated RF of the cloud albedo indirect effect. Peng and Lohmann  
37 (2003), Rotstain and Liu (2003) and Chen and Penner (2005) studied the sensitivity of their estimates to this  
38 dispersion effect. These studies confirm that their estimates of the cloud albedo RF, without taking the  
39 droplet spectra change into account, are overestimated by about 15 to 35%.  
40

41 The effects of aerosol particles on heterogeneous ice formation are currently insufficiently understood and  
42 present another level of challenge for both observations and modelling. Ice crystal concentrations cannot be  
43 easily measured with present *in situ* instrumentation because of the difficulty of detecting small particles  
44 (Hirst et al., 2001) and frequent shattering of ice particles when impacting the probes (Korolev and Isaac,  
45 2005). Current GCMs do not have sufficient rigor of the microphysics or sub-grid scale processes to  
46 accurately predict cirrus clouds or super-cooled clouds explicitly. Ice particles in clouds are often  
47 represented in by simple shapes (e.g., spheres), even though it is well known that few ice crystals are like  
48 that in reality. The radiative properties of ice particles in GCMs often do not effectively simulate the  
49 irregular shapes that are normally found, nor do they simulate the inclusions of crustal material or soot in the  
50 crystals.  
51

#### 52 2.4.5.6 Assessment of the Cloud Albedo RF

53

54 As in the TAR, only the aerosol interaction in the context of liquid water clouds is assessed, with knowledge  
55 of the interaction with ice clouds deemed insufficient. Since the TAR, the cloud albedo effect has been  
56 estimated in a more systematic way, and more modelling results are now available. Models now are more  
57 advanced in capturing the complexity of the aerosol-cloud interactions through forward computations. Even

1 though major uncertainties remain, clear progress has been made, leading to a convergence of the estimates  
2 from the different modelling efforts. Based on the results from all the modelling studies shown in Figure  
3 2.14, compared to the TAR it is now possible to present a best estimate for the cloud albedo RF of  $-0.7 \text{ W m}^{-2}$   
4 <sup>2</sup> as the median, with a 5–95% range of  $-0.3$  to  $-1.8 \text{ W m}^{-2}$ . The increase in the knowledge of the aerosol-  
5 cloud interactions and the reduction in the spread of the cloud albedo RF since the TAR result in an elevation  
6 of the level of scientific understanding to *low* (Table 2.11).

## 8 **2.5 Anthropogenic Changes in Surface Albedo and the Surface Energy Budget**

### 10 **2.5.1 Introduction**

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

31 Surface albedo can also be modified by the settling of anthropogenic aerosols on the ground, especially in  
32 the case of black carbon on snow (Hansen and Nazarenko, 2004). This mechanism may be considered to be  
33 an RF mechanism because diagnostic calculations may be performed under the strict definition of RF (see  
34 Sections 2.2 and 2.8). This mechanism was not discussed in the TAR.

36 Land cover change can also affect other physical properties such as surface emissivity, the fluxes of moisture  
37 through evaporation and transpiration, the ratio of latent to sensible heat fluxes (the Bowen ratio) and the  
38 aerodynamic roughness which exerts frictional drag on the atmosphere and also affects turbulent transfer of  
39 heat and moisture. All these processes can affect the air temperature near the ground and also modify  
40 humidity, precipitation and windspeed. Direct human perturbations to the water cycle, such as irrigation, can  
41 affect surface moisture fluxes and hence the surface energy balance. Changes in vegetation cover can affect  
42 the production of dust, which then exerts an RF. Changes in certain gases, particularly  $\text{CO}_2$  and  $\text{O}_3$ , can also  
43 exert an additional influence on climate by affecting the Bowen ratio, through plant responses which affect  
44 transpiration. These processes are discussed in detail in Chapter 7 (Section 7.2). While such processes will  
45 act as anthropogenic perturbations to the climate system (Pielke Sr. et al., 2002) and will fall at least partly  
46 within the “forcing” component of the forcing-feedback-response conceptual model, it is difficult to  
47 unequivocally quantify the pure forcing component as distinct from feedbacks and responses. The term  
48 “non-radiative forcing” has been proposed (Jacob et al., 2005) and this report adopts the similar term “non-  
49 initial-radiative effect”, but no quantitative metric separating forcing from feedback and response has yet  
50 been implemented for climatic perturbation processes which do not act directly on the radiation budget (see  
51 Section 2.2).

53 Energy consumption by human activities, such as heating of buildings, powering of electrical appliances and  
54 combustion of fuel by vehicles, can directly release heat into the environment. This was not discussed in the  
55 TAR. Anthropogenic heat release is not an RF, in that it does not directly perturb the radiation budget; the  
56 mechanisms are not well identified and so is here referred to as a non-initial-radiative effect. It can, however,  
57 be quantified as a direct input of energy to the system in terms of  $\text{W m}^{-2}$ .

### 2.5.2 *Changes in Land Cover Since 1750*

In 1750, 7.9–9.2 million km<sup>2</sup> (6%–7% of the global land surface) were under cultivation or pasture (Figure 2.15), 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. 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 and 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<sup>2</sup> (35%–39% of global land), and forest cover had decreased by roughly 11 million km<sup>2</sup> (Ramankutty and Foley, 1999; Klein Goldewijk, 2001) (Table 2.8).

[INSERT FIGURE 2.15 HERE]

Overall, most deforestation until the mid-20th Century occurred in the temperate regions (Figure 2.15). 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: Land-use*

Since the TAR, a number of estimates of the RF from land-use changes over the industrial era have been made (Table 2.8). Unlike the main TAR estimate, most of the more recent studies are “pure” radiative forcing calculations with the only change being land cover; feedbacks such as changes in snow cover are excluded. Brovkin et al. (2006) estimated the global mean RF relative to 1700 to be  $-0.15 \text{ W m}^{-2}$ , 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 RF relative to 1750 to be  $-0.15 \text{ W m}^{-2}$ . Using historical reconstructions of both croplands (Ramankutty and Foley, 1999) and pasturelands (Klein Goldewijk, 2001), Betts et al. (2006) simulated an RF of  $-0.18 \text{ W m}^{-2}$  since 1750. This study also estimated the RF relative to PNV to be  $-0.24 \text{ W m}^{-2}$ . 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 this RF as  $-0.08 \text{ W m}^{-2}$ . Myhre et al. (2005b) used land cover and albedo data from MODIS (Friedl et al., 2002; Schaaf et al., 2002) and estimated this RF as  $-0.09 \text{ W m}^{-2}$ . 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  $-5 \text{ W m}^{-2}$  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 2.15). In historical simulations, the spatial patterns of RF relative to the PNV 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 RF may depend on aerosol concentrations. Estimates of time evolution of aerosol RF and surface albedo RF may need to consider changes in each other (Betts et al., 2006).

### 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 RF estimates reported in the TAR used atlas-based datasets for present-day vegetation (Matthews, 1983; Wilson and 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  $-0.2 \text{ W m}^{-2}$  with the Wilson and Henderson-Sellers (1985) atlas-based land use dataset to  $-0.24 \text{ W m}^{-2}$  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., 2006). Myhre and Myhre (2003) found the RF relative to PNV to vary from  $-0.66 \text{ W m}^{-2}$  to  $0.29 \text{ W m}^{-2}$  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 the fraction of land under crops at  $0.5^\circ$  resolution from 1700 to 1990 (Figure 2.15, 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 2.15, 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  $0.5^\circ$  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 2.15) 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. Matthews et al. (2004) simulated RF relative to 1700 as  $-0.20 \text{ W m}^{-2}$  and  $-0.28 \text{ W m}^{-2}$  with the above land use reconstructions.

[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, 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., 2005b). 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  $-0.65 \text{ W m}^{-2}$  to  $+0.47 \text{ W m}^{-2}$ ; 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, the resulting RFs relative to PNV were  $-0.06$ ,  $-0.20$ , and  $-0.29 \text{ W m}^{-2}$  respectively. Similar results were found by Matthews et al. (2003) considering only cropland changes and not pasture; with cropland surface albedos of 0.17 and 0.20, RFs relative to 1700 were  $-0.15$  and  $-0.28 \text{ W m}^{-2}$  respectively.

1  
2 (iv) When climate models are used to estimate the RF, uncertainties in other parts of the model also affect  
3 the estimates. In particular, the simulation of snow cover affects the extent to which land cover changes  
4 affect surface albedo. Betts (2000) estimated that the systematic biases in snow cover in HadAM3 introduced  
5 errors of up to approximately 10% in the simulation of local RF due to conversion between forest and open  
6 land. Such uncertainties could be reduced by the use of an observational snow climatology in a model which  
7 just treats the radiative transfer (Myhre and Myhre, 2003). The simulation of cloud cover affects the extent to  
8 which the simulated surface albedo changes impact on planetary albedo – too much cloud cover could  
9 diminish the contribution of surface albedo changes to the planetary albedo change.

10  
11 On the basis of the studies assessed here, including a number of new estimates since the TAR, the  
12 assessment is that the best estimate of RF relative to 1750 due to land-use related surface albedo change  
13 should remain at  $-0.2 \pm 0.2 \text{ W m}^{-2}$ . In the light of the additional modelling studies, the exclusion of  
14 feedbacks, the improved incorporation of large-scale observations, and the explicit consideration of land use  
15 reconstructions for 1750, the level of scientific understanding is raised to *medium-low*, compared to *low* in  
16 the TAR (Table 2.11).

#### 17 18 **2.5.4 Radiative Forcing by Anthropogenic Surface Albedo Change: Black Carbon in Snow and Ice**

19  
20 The presence of soot particles in snow could cause a decrease in the albedo of snow and affect snowmelt.  
21 Initial estimates by Hansen et al. (2000) suggested that black carbon could thereby exert a positive RF of  
22  $+0.2 \text{ W m}^{-2}$ . This estimate was refined by Hansen and Nazarenko (2004) who used measured BC  
23 concentrations within snow/ice at a wide range of geographic locations to deduce the perturbation to the  
24 surface and planetary albedo, deriving an RF of  $+0.15 \text{ W m}^{-2}$ . The uncertainty in this estimate is substantial  
25 due to uncertainties in: (i) whether BC and snow particles are internally or externally mixed; (ii) BC and  
26 snow particle shapes and sizes; (iii) voids within BC particles; (iv) uncertainties in the BC imaginary  
27 refractive index. Jacobson (2004) developed a global model that allows the BC aerosol to enter snow via  
28 precipitation and dry deposition, thereby modifying the snow albedo and emissivity. They found modelled  
29 concentrations of BC within snow that were in reasonable agreement with those from many observations.  
30 The model study found that black carbon on snow and sea ice caused a decrease in the surface albedo of  
31 0.4% globally and 1% in the northern hemisphere, although RFs were not reported. Hansen et al. (2005)  
32 allowed the albedo change to be proportional to local BC deposition according to Koch (2001) and presented  
33 a further revised estimate of  $0.08 \text{ W m}^{-2}$ . They also suggested that this RF mechanism produces a greater  
34 temperature response by a factor of 1.7 than an equivalent  $\text{CO}_2$  RF i.e., the ‘efficacy’ may be higher for this  
35 RF mechanism (see Section 2.8.5.7). This report adopts a best estimate for the black carbon on snow RF of  
36  $+0.10 \pm 0.10 \text{ W m}^{-2}$ , with a *low* level of scientific understanding (Table 2.11).

#### 37 38 **2.5.5 Other Effects of Anthropogenic Changes in Land Cover**

39  
40 Anthropogenic land use and land cover change can also modify climate through other mechanisms, some  
41 directly perturbing the Earth radiation budget and some perturbing other processes. Impacts of land cover  
42 change on emissions of  $\text{CO}_2$ ,  $\text{CH}_4$ , biomass burning aerosols and dust aerosols are discussed in Sections 2.3  
43 and 2.4. Land cover change itself can also modify the surface energy and moisture budgets through changes  
44 in evaporation and the fluxes of latent and sensible heat, directly affecting precipitation and atmospheric  
45 circulation as well as temperature. Model results suggest that the combined effects of past tropical  
46 deforestation may have exerted regional warmings of approximately 0.2 K relative to PNV, and may have  
47 perturbed the global atmospheric circulation affecting regional climates remote from the land cover change  
48 (Chase et al., 2000; Zhao et al., 2001; Pielke Sr. et al., 2002, Chapters 7, 9 and 11).

49  
50 Since the dominant aspect of land cover change since 1750 has been deforestation in temperate regions, the  
51 overall effect of anthropogenic land cover change on global temperature will depend largely on the relative  
52 importance of increased surface albedo in winter and spring (exerting a cooling) and reduced evaporation in  
53 summer and in the tropics (exerting a warming) (Bounoua et al., 2002). Estimates of global temperature  
54 responses from past deforestation vary from 0.01 K (Zhao et al., 2001) to  $-0.25 \text{ K}$  (Govindasamy et al.,  
55 2001a; Brovkin et al., 2006). If cooling by increased surface albedo dominates, then the historical effect of  
56 land cover change may still be adequately represented by RF. With tropical deforestation becoming more  
57 significant in recent decades, warming due to reduced evaporation may become more significant globally

1 than increased surface albedo. RF would then be less useful as a metric of climate change induced by land  
2 cover change recently and in the future.

### 3 4 **2.5.6 Tropospheric Water Vapour from Anthropogenic Sources**

5  
6 Anthropogenic use of water is less than 1% of natural sources of water vapour and about 70% of the use of  
7 water for human activity is from irrigation (Döll, 2002). Several regional studies have indicated an impact of  
8 irrigation on temperature, humidity, and precipitation (Barnston and Schickedanz, 1984; Lohar and Pal,  
9 1995; de Ridder and Gallée, 1998; Moore and Rojstaczer, 2001; Zhang et al., 2002). Boucher et al. (2004)  
10 used a GCM to show that irrigation has a global impact on temperature and humidity. Over Asia where most  
11 of the irrigation takes place the simulations showed a change in the water vapour content in the lower  
12 troposphere by up to 1%, resulting in an RF of  $0.03 \text{ W m}^{-2}$ . However, the effect of irrigation on surface  
13 temperature was dominated by evaporative cooling rather than by the excess greenhouse effect and thus a  
14 decrease in surface temperature was found. Since irrigation impacts the temperature, humidity, clouds, and  
15 precipitation as well as the natural evaporation through changes in the surface temperature, this questions the  
16 strict use of RF. Uncertainties in the water vapour flow to the atmosphere from irrigation are significant and  
17 Gordon et al. (2005) give a substantially higher estimate compared to that of Boucher et al. (2004). Most of  
18 this uncertainty is likely to be linked to differences between the total withdrawal for irrigation and the  
19 amount actually used (Boucher et al., 2004). Furthermore, Gordon et al. (2005) also estimate a reduced water  
20 vapour flow to the atmosphere from deforestation, most importantly in tropical areas. This reduced water  
21 vapour flow is a factor of three larger than the water vapour increase due to irrigation in Boucher et al.  
22 (2004), but so far there are no estimates of the effect of this on the water vapour content of the atmosphere  
23 and its RF. Water vapour changes from deforestation will, like irrigation, impact the surface evaporation and  
24 temperature as well as impact the water cycle in the atmosphere. RF from anthropogenic sources of  
25 tropospheric water vapour is not evaluated here, since these sources impact surface temperature more  
26 significantly through these non-radiative processes, and a strict use of the RF is problematic. The emission of  
27 water vapour from fossil fuel combustion is significantly lower than the emission from changes in land use  
28 (Boucher et al., 2004).

### 29 30 **2.5.7 Anthropogenic Heat Release**

31  
32 Urban heat islands result partly from the physical properties of the urban landscape and partly from the  
33 release of heat into the environment by the use of energy by human activities such as the heating of buildings  
34 and the powering of appliances and vehicles (Human Energy Production). The global total heat flux from  
35 this is estimated as  $0.03 \text{ W m}^{-2}$  (Nakicenovic, 1998). If this energy release were concentrated in cities, which  
36 are estimated to cover 0.046% of the Earth's surface (Loveland et al., 2000) the mean local heat flux in a city  
37 would be  $65 \text{ W m}^{-2}$ . Daytime values in central Tokyo typically exceed  $400 \text{ W m}^{-2}$  with a maximum of  $1590$   
38  $\text{W m}^{-2}$  in winter (Ichinose et al., 1999). Although Human Energy Production is a small influence at the  
39 global scale, it may be very important for climate changes in cities (Betts and Best, 2004; Crutzen, 2004).

### 40 41 **2.5.8 Effects of CO<sub>2</sub> Changes on Climate via Plant Physiology: "Physiological Forcing"**

42  
43 As well as exerting an RF on the climate system, increasing concentrations of atmospheric CO<sub>2</sub> can also  
44 perturb the climate system through direct effects on plant physiology. Plant stomatal apertures open less  
45 under higher CO<sub>2</sub> concentrations (Field et al., 1995), which directly reduces the flux of moisture from the  
46 surface to the atmosphere through transpiration (Sellers et al., 1996). A decrease in moisture flux modifies  
47 the surface energy balance, increasing the ratio of sensible heat flux to latent heat flux and therefore warming  
48 the air near the surface (Sellers et al., 1996; Betts et al., 1997; Cox et al., 1999). Betts et al. (2004) propose  
49 the term "*physiological forcing*" for this mechanism. Although no studies have yet explicitly quantified the  
50 temperature response to *physiological forcing* at the present-day, the presence of this forcing has been  
51 detected in global hydrological budgets (Gedney et al., 2006, Chapter 9: Section 9.5). This process can be  
52 considered a non-initial-radiative effect, as distinct from a feedback, since the mechanism involves a direct  
53 response to CO<sub>2</sub> rise and not a response to climate change. It is not possible to quantify this with RF.  
54 Reduced global transpiration would also be expected to reduce atmospheric water vapour causing a negative  
55 forcing, but no estimates of this have been made.

1 Increased CO<sub>2</sub> concentrations can also fertilize photosynthesis, which models suggest will have contributed  
2 to increased vegetation cover and leaf area over the 20th Century (Cramer et al., 2001). Vegetation Index, a  
3 remote sensing product indicative of leaf area, biomass and potential photosynthesis, has been observed to  
4 increase (Zhou et al., 2001) although other causes including climate change itself are also *likely* to have  
5 contributed. Increased vegetation cover and leaf area would decrease surface albedo, which would act to  
6 oppose the increase in albedo due to deforestation. The RF due to this process has not been evaluated and  
7 there is a *very low* scientific understanding of these effects.

## 8 9 **2.6 Contrails and Aircraft-Induced Cloudiness**

### 10 **2.6.1 Introduction**

11 The IPCC separately evaluated the RF from subsonic and supersonic aircraft operations in the Special Report  
12 on Aviation and the Global Atmosphere (IPCC, 1999), hereinafter designated as IPCC-1999. Like many  
13 other sectors, subsonic aircraft operations around the globe contribute directly and indirectly to the RF of  
14 climate change. This section only assesses the aspects that are unique to the aviation sector, namely the  
15 formation of persistent condensation trails (contrails), their impact on cirrus cloudiness, and the effects of  
16 aviation aerosols. Persistent contrail formation and induced cloudiness are indirect effects from aircraft  
17 operations because they depend on variable humidity and temperature conditions along aircraft flight tracks.  
18 Thus, future changes in atmospheric humidity and temperature distributions in the upper troposphere will  
19 have consequences for aviation-induced cloudiness. Also noted here is the potential role of aviation aerosols  
20 in altering the properties of clouds that form later in air containing aircraft emissions.

### 21 **2.6.2 Radiative Forcing Estimates for Persistent Line-Shaped Contrails**

22 Aircraft produce persistent contrails in the upper troposphere in ice-supersaturated air masses (IPCC, 1999).  
23 Contrails are thin cirrus clouds, which reflect solar radiation and trap outgoing long-wave radiation. The  
24 latter effect is expected to dominate for thin cirrus (Hartmann et al., 1992; Meerkötter et al., 1999), thereby  
25 resulting in a net positive RF value for contrails. Persistent contrail cover has been calculated globally from  
26 meteorological data (e.g., Sausen et al., 1998) or by using a modified cirrus cloud parameterization in a  
27 GCM (Ponater et al., 2002). Contrail cover calculations are uncertain because the extent of supersaturated  
28 regions in the atmosphere is poorly known. The associated contrail RF follows from determining an optical  
29 depth for the computed contrail cover. The global RF values for contrail and induced cloudiness are assumed  
30 to vary linearly with distances flown by the global fleet if flight ambient conditions remain unchanged. The  
31 current best estimate for the RF of persistent linear contrails for aircraft operations in 2000 is 0.010 W m<sup>-2</sup>  
32 (Table 2.9; Sausen et al., 2005). The value is based on independent estimates derived from Myhre and  
33 Stordal (2001b) and Marquart et al. (2003) that were updated for increased aircraft traffic in Sausen et al.  
34 (2005) to give RF estimates of 0.015 W m<sup>-2</sup> 0.006 W m<sup>-2</sup> respectively. The uncertainty range is  
35 conservatively estimated to be a factor of 3. The 0.010 W m<sup>-2</sup> value is also considered to be the best estimate  
36 for 2004 because of the slow overall growth in aviation fuel use in the 2000–2004 period. The decrease in  
37 the best estimate from the TAR by a factor of 2 results from reassessments of persistent contrail cover and  
38 lower optical depth estimates (Marquart and Mayer, 2002; Meyer et al., 2002; Ponater et al., 2002; Marquart  
39 et al., 2003). The new estimates include diurnal changes in the solar RF, which decreases the net RF for a  
40 given contrail cover by about 20% (Myhre and Stordal, 2001b). The level of scientific understanding of  
41 contrail RF is considered *low*, since important uncertainties remain in the determination of global values  
42 (Table 2.11). For example, unexplained regional differences are found in contrail optical depths between  
43 Europe and the United States that have not been fully accounted for in model calculations (Palikonda et al.,  
44 2005; Meyer et al., 2002; Ponater et al., 2002).

45 [INSERT TABLE 2.9 HERE]

### 46 **2.6.3 Radiative Forcing Estimates for Aviation-Induced Cloudiness**

47 Individual persistent contrails are routinely observed to shear and spread, covering large additional areas  
48 with cirrus cloud (Minnis et al., 1998). Aviation aerosol could also lead to changes in cirrus cloud (see  
49 Section 2.6.4). Aviation-induced cloudiness (AIC) is defined to be the sum of all changes in cloudiness  
50 associated with aviation operations. Thus, an AIC estimate includes persistent contrail cover. Because

1 spreading contrails lose their characteristic linear shape, a component of AIC is indistinguishable from  
2 background cirrus. This basic ambiguity, which prevented the formulation of a best estimate of AIC amounts  
3 and the associated RF in IPCC-1999, still exists for this assessment. Estimates of the ratio of induced  
4 cloudiness cover to that of persistent linear contrails range from 1.8 to 10 (Minnis et al., 2004; Mannstein  
5 and Schumann, 2005), indicating the uncertainty in estimating AIC amounts. First attempts to quantify AIC  
6 used trend differences in cirrus cloudiness between regions of high aviation fuel consumption and low  
7 consumption (Boucher, 1999). Since IPCC-1999, two studies have also found significant positive trends in  
8 cirrus cloudiness in some regions of high air traffic and found lower to negative trends outside air traffic  
9 regions (Zerefos et al., 2003; Stordal et al., 2005). Using the International Satellite Cloud Climatology  
10 Project (ISCCP) database, these studies derived cirrus cover trends for Europe of 1 to 2% per decade over  
11 the last one to two decades. A study with the TOVS satellite provides further support for these trends  
12 (Stubenrauch and Schumann, 2005). However, cirrus trends that occurred due to natural variability, climate  
13 change, or other anthropogenic effects could not be accounted for in these studies. Cirrus trends over the  
14 United States but not over Europe were found to be consistent with changes in contrail cover and frequency  
15 (Minnis et al., 2004). Thus, significant uncertainty remains in attributing observed cirrus trends to aviation.  
16

17 Regional cirrus trends were used as a basis to compute a global mean RF value for AIC in 2000 of  $0.030 \text{ W m}^{-2}$   
18  $\text{m}^{-2}$  with a range of  $0.01\text{--}0.08 \text{ W m}^{-2}$  (Stordal et al., 2005). This value is not considered a best estimate  
19 because of the uncertainty in the optical properties of AIC and in the assumptions used to derive AIC cover.  
20 However, this value is in good agreement with the upper limit estimate for AIC RF in 1992 of  $0.026 \text{ W m}^{-2}$   
21 derived from surface and satellite cloudiness observations (Minnis et al., 2004). A value of  $0.03 \text{ W m}^{-2}$  is  
22 close to the upper limit estimate of  $0.04 \text{ W m}^{-2}$  derived for non-contrail cloudiness in IPCC-1999. Without  
23 an AIC best estimate, the best estimate of the total RF value for aviation-induced cloudiness (Table 2.12 and  
24 Figure 2.20) includes only that due to persistent linear contrails. RF estimates for AIC made using cirrus  
25 trend data necessarily cannot distinguish between the components of aviation cloudiness, namely persistent  
26 linear contrails, spreading contrails, and other aviation aerosol effects. Some aviation effects might be more  
27 appropriately considered feedback processes rather than an RF (see Sections 2.2 and 2.4.5). However, our  
28 low understanding of the processes involved and the lack of quantitative approaches preclude reliably  
29 making the forcing/feedback distinction for all aviation effects in this assessment.  
30

31 Two issues related to the climate response of aviation cloudiness are worth noting here. First, Minnis et al.  
32 (2004; 2005) use their RF estimate for total aviation-induced cloudiness over the USA in an empirical model  
33 to conclude that the surface temperature response for the period 1973–1994 could be as large as the observed  
34 surface warming over the USA (around 0.3 K per decade). In response to the Minnis et al. conclusion,  
35 contrail RF was examined in two global climate-modelling studies (Hansen et al., 2005; Ponater et al., 2005).  
36 Both studies concluded that the surface temperature response calculated by Minnis et al. (2004) is too large  
37 by one to two orders of magnitude. For the Minnis et al. result to be correct, the climate efficacy or climate  
38 sensitivity of contrail RF would need to be much greater than that of other larger RF terms, e.g.,  $\text{CO}_2$ .  
39 Instead, contrail RF is found to have a smaller efficacy than an equivalent  $\text{CO}_2$  RF (Hansen et al., 2005;  
40 Ponater et al., 2005) (see Section 2.8.5.7), which is consistent with the general ineffectiveness of high clouds  
41 in influencing diurnal surface temperatures (Hansen et al., 1995; 2005). Several substantive explanations for  
42 the incorrectness of the enhanced response found in the Minnis et al. study have been presented (Hansen et  
43 al., 2005; Ponater et al., 2005; Shine, 2005).  
44

45 The second issue is that the absence of aviation-induced cloudiness has been proposed as the cause of the  
46 increased diurnal temperature range (DTR) found in surface observations made during the short period when  
47 all USA air traffic was grounded starting on 11 September 2001 (Travis et al., 2002; Travis et al., 2004). The  
48 Travis et al. studies show that during this period: (i) DTR was enhanced across the conterminous USA, with  
49 increases in the maximum temperatures that were not matched by increases of similar magnitude in the  
50 minimum temperatures, and (ii) the largest DTR changes corresponded to regions with the greatest contrail  
51 cover. The Travis et al. conclusions are weak because they are based on a correlation rather than a  
52 quantitative model and rely (necessarily) on very limited data (Schumann, 2005). Unusually clear weather  
53 across the USA during the shutdown period also has been proposed to account for the observed DTR  
54 changes (Kalkstein and Balling Jr., 2004). Thus, more evidence and a quantitative physical model are needed  
55 before the validity of the proposed relationship between regional contrail cover and DTR could be  
56 considered further.  
57

## 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 modelling 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 as well as in regions far away from flight corridors because of aerosol transport. 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 reduced their effectiveness as ice nuclei (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-cloud interactions discussed in Sections 2.4.1 and 2.4.5, 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

The estimates of long-term solar irradiance changes used in the TAR (e.g., Hoyt and Schatten, 1993; Lean et al., 1995) have been revised downwards, based on new studies indicating that bright solar faculae *likely* contribute a smaller irradiance increase since the Maunder Minimum than was originally suggested by the range of brightness in Sun-like stars (Hall and Lockwood, 2004; M. Wang et al., 2005). However, empirical results since the TAR have strengthened the evidence for solar forcing of climate change by identifying detectable tropospheric changes associated with solar variability, including during the solar cycle (Chapter 9, Section 9.2, van Loon and Shea, 2000; Douglass and Clader, 2002; Gleisner and Thejll, 2003; Haigh, 2003; White et al., 2003; Stott et al., 2003; Coughlin and Tung, 2004; Labitzke, 2004; Crooks and Gray, 2005). The most likely mechanism is considered to be some combination of direct forcing by changes in total solar irradiance, and indirect effects of UV radiation on the stratosphere. Least certain, and under ongoing debate as discussed in the TAR, are indirect effects induced by galactic cosmic rays (e.g., Marsh and Svensmark, 2000a, 2000b, Kristjánsson et al., 2002, Sun and Bradley, 2002).

#### 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, contributing to a database extant 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 2.16, 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

1 radiometer prior to determining radiometric offsets. In contrast, the ACRIM composite (Willson and  
2 Mordvinov, 2003), also shown in Figure 2.16, utilizes ACRIMSAT rather than VIRGO observations in  
3 recent times and cross calibrates the reported data assuming that radiometric sensitivity drifts have already  
4 been fully accounted for. A third composite, the Space Absolute Radiometric Reference (SARR) composite,  
5 uses individual absolute irradiance measurements from the shuttle to cross calibrate satellite records (Dewitte  
6 et al., 2005). The gross temporal features of the composite irradiance records are very similar, each showing  
7 day-to-week variations associated with the Sun's rotation on its axis, and decadal fluctuations arising from  
8 the 11-year solar activity cycle. But the linear slopes differ among the three different composite records, as  
9 do levels at solar activity minima (1986 and 1996). These differences are the result of different cross  
10 calibrations and drift adjustments applied to individual radiometric sensitivities when constructing the  
11 composites (Fröhlich and Lean, 2004).

12  
13 [INSERT FIGURE 2.16 HERE]  
14

15 Solar irradiance levels are comparable in the two most recent cycle minima when absolute uncertainties and  
16 sensitivity drifts in the measurements are assessed (Fröhlich and Lean, 2004, and references therein). The  
17 increase in excess of 0.04% over the 27-year period of the ACRIM irradiance composite (Willson and  
18 Mordvinov, 2003), though incompletely understood, is thought to be more of instrumental rather than solar  
19 origin (Fröhlich and Lean, 2004). The irradiance increase in the ACRIM composite is indicative of an  
20 episodic increase between 1989 and 1992 that is present in the Nimbus 7 data (Lee et al., 1995; Chapman et  
21 al., 1996). Independent, overlapping ERBS observations do not show this increase; nor do they suggest a  
22 significant secular trend (Lee et al., 1995). Such a trend is not present in the PMOD composite, in which  
23 total irradiance between successive solar minima is nearly constant, to better than 0.01% (Fröhlich and Lean,  
24 2004). Although a long-term trend of order 0.01% is present in the SARR composite between successive  
25 solar activity minima (in 1986 and 1996), it is not statistically significant because the estimated uncertainty  
26 is  $\pm 0.026\%$  (Dewitte et al., 2005).

27  
28 Current understanding of solar activity and the known sources of irradiance variability suggests comparable  
29 irradiance levels during the past two solar minima. The primary known cause of contemporary irradiance  
30 variability is the presence on the Sun's disk of sunspots (compact, dark features where radiation is locally  
31 depleted) and faculae (extended bright features where radiation is locally enhanced). Models that combine  
32 records of the global sunspot darkening calculated directly from white light images and the Mg irradiance  
33 index as a proxy for the facular signal do not exhibit a significant secular trend during activity minima  
34 (Fröhlich and Lean, 2004; Preminger and Walton, 2005). Nor do the modern instrumental measurements of  
35 galactic cosmic rays, 10.7 cm flux and the aa-index since the 1950s (Benestad, 2005) indicate this feature.  
36 While changes in surface emissivity by magnetic sunspot and facular regions are, from a theoretical view,  
37 the most effective in altering irradiance (Spruit, 2000), other mechanisms have also been proposed that may  
38 cause additional, possibly secular, irradiance changes. Of these, changes in solar diameter have been  
39 considered a likely candidate (e.g., Sofia and Li, 2001). But recent analysis of solar imagery, primarily from  
40 the Michelson Doppler Imager (MDI) instrument on SOHO, indicates that solar diameter changes are no  
41 more than a few km per year during the solar cycle (Dziembowski et al., 2001), for which associated  
42 irradiance changes are 0.001%, two orders of magnitude less than the measured solar irradiance cycle.

#### 43 44 2.7.1.1.3 *Measurements of solar spectral irradiance*

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

53  
54 Radiation in the visible and IR spectrum has a notably different temporal character than the spectrum below  
55 300 nm. Maximum energy changes occur at wavelengths from 400 to 500 nm. Fractional changes are  
56 greatest at UV wavelengths but the actual energy change is considerably smaller than in the visible spectrum.

1 Over the time scale of the 11-year solar cycle, bolometric facular brightness exceeds sunspot blocking by  
2 about a factor of two, and there is an increase in spectral irradiance at most, if not all, wavelengths from the  
3 minimum to the maximum of the solar cycle. Estimated solar cycle changes are 0.08% in the total solar  
4 irradiance. Broken down by wavelength range these irradiance changes are 1.3% at 200–300 nm, 0.2% at  
5 315–400 nm, 0.08% at 400–700 nm, 0.04% at 700–1000 nm and 0.025% at 1000–1600 nm.

6 However, during episodes of strong solar activity sunspot blocking can dominate facular brightening,  
7 causing decreased irradiance at most wavelengths. Spectral irradiance changes on these shorter time scales  
8 now being measured by SORCE provide tests of the wavelength-dependent sunspot and facular  
9 parameterizations in solar irradiance variability models. The modelled spectral irradiance changes are in  
10 good overall agreement with initial SORCE observations but as yet the SORCE observations are too short to  
11 provide definitive information about the amplitude of solar spectral irradiance changes during the solar  
12 cycle.

### 14 2.7.1.2 *Estimating Past Solar Radiative Forcing*

#### 16 2.7.1.2.1 *Reconstructions of past variations in solar irradiance*

17 Long-term solar irradiance changes over the past four hundred years may be less by a factor of 2 to 4 than in  
18 the reconstructions employed by the TAR for climate change simulations. Irradiance reconstructions such as  
19 those of Hoyt and Schatten (1993), Lean et al. (1995), Lean (2000), Lockwood and Stamper (1999) and  
20 Solanki and Fligge (1999), used in the TAR, assumed the existence of a long-term variability component in  
21 addition to the known 11-year cycle, in which the seventeenth century Maunder Minimum total irradiance  
22 was reduced in the range of 0.15% to 0.3% below contemporary solar minima. The temporal structure of this  
23 long-term component, typically associated with facular evolution, was assumed to track either the smoothed  
24 amplitude of the solar activity cycle or the cycle length. The motivation for adopting a long-term irradiance  
25 component was three-fold. Firstly, the range of variability in Sun-like stars (Baliunas and Jastrow, 1990)  
26 secondly, the long-term trend in geomagnetic activity and, thirdly, solar modulation of cosmogenic isotopes,  
27 all suggested that the Sun is capable of a broader range of activity than witnessed during recent solar cycles  
28 (i.e., the observational record in Figure. 2.16). Various estimates of the increase in total solar irradiance from  
29 the seventeenth century Maunder Minimum to the current activity minima from these irradiance  
30 reconstructions are compared with recent results in Table 2.10.

32 [INSERT TABLE 2.10 HERE]

34 Each of the above three assumptions for the existence of a significant long-term irradiance component is now  
35 questionable. A reassessment of the stellar data has been unable to recover the original bimodal separation of  
36 (lower) Ca emission in non-cycling stars (assumed to be in Maunder Minimum type states) compared with  
37 (higher) emission in cycling stars (Hall and Lockwood, 2004) which underpins the Lean et al. (1995) and  
38 Lean (2000) irradiance reconstructions. Rather, the current Sun is thought to have “typical” (rather than  
39 high) activity relative to other stars. Plausible lowest brightness levels inferred from stellar observations are  
40 higher than the peak of the lower mode of the initial distribution of Baliunas and Jastrow (1990). Other  
41 studies raise the possibility of long-term instrumental drifts in historical indices of geomagnetic activity  
42 (Svalgaard et al., 2004), which would reduce somewhat the long-term trend in the Lockwood and Stamper  
43 (1999) irradiance reconstruction. Furthermore, the relationship between solar irradiance and geomagnetic  
44 and cosmogenic indices is complex, and not necessarily linear. Simulations of the transport of magnetic flux  
45 on the Sun and propagation of open flux into the heliosphere indicate that “open” magnetic flux (which  
46 modulates geomagnetic activity and cosmogenic isotopes) can accumulate on inter-cycle time scales even  
47 when closed flux (such as in sunspots and faculae) does not (Lean et al., 2002; Y. Wang et al., 2005).

49 A new reconstruction of solar irradiance based on a model of solar magnetic flux variations (Y. Wang et al.,  
50 2005), that does not invoke geomagnetic, cosmogenic or stellar proxies, suggests that the amplitude of the  
51 background component is significantly less than previously assumed, specifically 0.27 times that of Lean  
52 (2000). This estimate results from simulations of the eruption, transport and accumulation of magnetic flux  
53 during the past 300 years using a flux transport model with variable meridional flow. Variations in both the  
54 total flux and in just the flux that extends into the heliosphere (the open flux) are estimated, arising from the  
55 deposition of bipolar magnetic regions (active regions) and smaller-scale bright features (ephemeral regions)  
56 on the Sun’s surface, in strengths and numbers proportional to the sunspot number. The open flux compares  
57 reasonably well with the cosmogenic isotopes whose variations arise, in part, from heliospheric modulation.

1 This gives confidence that the approach is plausible. A small accumulation of total flux (and possibly  
2 ephemeral regions) produces a net increase in facular brightness which, in combination with sunspot  
3 blocking, permits the reconstruction of total solar irradiance shown in Figure 2.17. There is a 0.04% increase  
4 from the Maunder Minimum to present-day cycle minima.

5  
6 [INSERT FIGURE 2.17 HERE]  
7

8 Prior to direct telescopic measurements of sunspots, which commenced around 1610, knowledge of solar  
9 activity is inferred indirectly from the <sup>14</sup>C and <sup>10</sup>Be cosmogenic isotope records in tree-rings and ice-cores,  
10 respectively, which exhibit solar-related cycles near 90-, 200- and 2300 years. Some studies of cosmogenic  
11 isotopes (Jirikowic and Damon, 1994) and spectral analysis of the sunspot record (Rigozo et al., 2001)  
12 suggest that solar activity during the twelfth century Medieval Solar Maximum was comparable to the  
13 present Modern Solar Maximum. Recent work attempts to account for the chain of physical processes in  
14 which solar magnetic fields modulate the heliosphere, in turn altering the penetration of the galactic cosmic  
15 rays whose flux produces the cosmogenic isotopes which are subsequently deposited in the terrestrial system  
16 following additional transport and chemical processes. An initial effort reported exceptionally high levels of  
17 solar activity in the past 70 years, relative to the preceding eight thousand years (Solanki et al., 2004). In  
18 contrast, when differences among isotopes records are taken into account and the <sup>14</sup>C record corrected for  
19 fossil fuel burning, current levels of solar activity are found to be historically high, but not exceptionally so  
20 (Muscheler et al., 2006).

#### 21 2.7.1.2.2 *Implications for solar radiative forcing*

22 In terms of plausible physical understanding, the most likely secular increase in total irradiance from the  
23 Maunder Minimum to current cycle minima is 0.04% (an irradiance increase of roughly 0.5 W m<sup>-2</sup> in 1365  
24 W m<sup>-2</sup>), corresponding to an RF<sup>11</sup> of 0.1 W m<sup>-2</sup>. The larger RF estimates in Table 2.10, in the range 0.38 to  
25 0.68 W m<sup>-2</sup>, correspond to assumed changes in solar irradiance at cycle minima derived from brightness  
26 fluctuations in Sun-like stars that are no longer valid. Since the 11-year cycle amplitude has increased from  
27 the Maunder Minimum to the present, the total irradiance increase to the present-day cycle mean is 0.08%.  
28 From 1750 to the present there is a net 0.05% increase in total solar irradiance, according to the 11-year  
29 smoothed total solar irradiance time series of Y. Wang et al. (2005), shown in Figure 2.17. This corresponds  
30 to an RF of 0.12 W m<sup>-2</sup>, which is more than a factor of two less than the solar RF estimate in the TAR, also  
31 from 1750 to the present. Using the Lean (2000) reconstruction (the lower envelope in Figure 2.17) as an  
32 upper limit, there is a 0.12% irradiance increase since 1750, for which the RF is 0.3 W m<sup>-2</sup>. The lower limit  
33 of the irradiance increase from 1750 to the present is 0.026% due to the increase in the 11-year cycle only.  
34 The corresponding lower limit of the RF is 0.06 W m<sup>-2</sup>. As with solar cycle changes, long-term irradiance  
35 variations are expected to have significant spectral dependence. For example, the Y. Wang et al. (2005) flux  
36 transport estimates imply decreases during the Maunder Minimum relative to contemporary activity cycle  
37 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  
38 0.02% at 1000 to 1600 nm (Lean et al., 2005), compared with, respectively, 1.4%, 0.32%, 0.17%, 0.1% and  
39 0.06% in the earlier model of Lean (2000).

#### 40 2.7.1.3 *Indirect Effects of Solar Variability*

41  
42 Approximately 1% of the Sun's radiant energy is in the ultraviolet portion of the spectrum at wavelengths  
43 below about 300 nm, which the Earth's atmosphere absorbs. Although of considerably smaller absolute  
44 energy than the total irradiance, solar UV radiation is fractionally more variable by at least an order of  
45 magnitude. It contributes significantly to changes in total solar irradiance (15% of the total irradiance cycle,  
46 Lean et al., 1997), creates and modifies the ozone layer, but is not considered as a direct RF because it does  
47 not reach the troposphere. Since the TAR, new studies have confirmed and advanced the plausibility of  
48  
49

---

<sup>11</sup> To estimate RF, the change in total solar irradiance is multiplied by a quarter to account for Earth-Sun geometry and then multiplied by 0.7 to account for the planetary albedo (e.g., Ramaswamy et al., 2001). Ideally this resulting RF should also be reduced by 15% to account for solar variations in the UV below 300nm (see Section 2.7.1.3) and further reduced by about 4% to account for stratospheric absorption of solar radiation above 300 nm and the resulting stratospheric adjustment (Hansen et al., 1997). However, these corrections are not made to the RF estimates in this report because they: 1) represent small adjustments to the RF; 2) may in part be compensated by indirect effects of solar-ozone interaction in the stratosphere (see Section 2.7.1.3); 3) are not routinely reported in the literature.

1 indirect effects involving the modification of the stratosphere by solar UV irradiance variations (and possibly  
2 by solar-induced variations in the overlying mesosphere and lower thermosphere), with subsequent  
3 dynamical and radiative coupling to the troposphere (Chapter 9, Section 9.2). Whether solar wind  
4 fluctuations (Boberg and Lundstedt, 2002) or solar-induced heliospheric modulation of galactic cosmic rays  
5 (Marsh and Svensmark, 2000b) also contribute indirect forcings remains ambiguous.

6  
7 As in the troposphere, anthropogenic effects, internal cycles (e.g., the Quasi Biennial Oscillation) and natural  
8 influences all affect the stratosphere. It is now well established from both empirical and model studies that  
9 solar cycle changes in UV radiation alter middle atmospheric ozone concentrations (Fioletov et al., 2002;  
10 Geller and Smyshlyaev, 2002; Hood, 2003), temperatures and winds (Ramaswamy et al., 2001; Labitzke et  
11 al., 2002; Haigh, 2003; Labitzke, 2004; Crooks and Gray, 2005) including the Quasi Biennial Oscillation  
12 (McCormack, 2003; Salby and Callaghan, 2004). In their recent survey of solar influences on climate, Gray  
13 et al. (2005) note that updated observational analyses have confirmed earlier 11-year cycle signals in zonally  
14 averaged stratospheric temperature, ozone and circulation with increased statistical confidence. There is a  
15 solar-cycle induced increase in global total ozone of 2–3% at solar cycle maximum, accompanied by  
16 temperature responses that increase with altitude, exceeding 1 K around 50 km. However, the amplitudes and  
17 geographical and altitudinal patterns of these variations are only approximately known, and are not linked in  
18 an easily discernible manner to the forcing. For example, solar forcing appears to induce a significant lower  
19 stratospheric response (Hood, 2003), which may have a dynamical origin caused by changes in temperature  
20 affecting planetary wave propagation, but it is not currently reproduced by models.

21  
22 When solar activity is high, the more complex magnetic configuration of the heliosphere reduces the flux of  
23 galactic cosmic rays in the Earth's atmosphere. Various scenarios have been proposed whereby solar-  
24 induced galactic cosmic ray fluctuations might influence climate (as surveyed by Gray et al., 2005). Carslaw  
25 et al. (2002) suggest that since the plasma produced by cosmic ray ionization in the troposphere is part of an  
26 electric circuit that extends from the Earth's surface to the ionosphere, cosmic rays may affect thunderstorm  
27 electrification. By altering the population of cloud condensation nuclei and hence microphysical cloud  
28 properties (droplet number and concentration) cosmic rays may also induce processes analogous to the  
29 indirect effect of tropospheric aerosols. The presence of ions, such as produced by cosmic rays, is recognized  
30 as influencing several microphysical mechanisms (Harrison and Carslaw, 2003) Aerosols may nucleate  
31 preferentially on atmospheric cluster ions. In case of low gas-phase sulphuric acid concentrations, ion-  
32 induced nucleation may dominate over binary sulphuric acid-water nucleation. Also, increased ion  
33 nucleation and increased scavenging rates of aerosols in turbulent regions around clouds seem likely.  
34 Because of the difficulty in tracking the influence of one particular modification brought about by ions  
35 through the long chain of complex interacting processes, quantitative estimates of galactic cosmic ray-  
36 induced changes in aerosol and cloud formation have not been reached.

37  
38 Many empirical associations have been reported between globally averaged low-level cloud cover and  
39 cosmic ray fluxes (e.g., Marsh and Svensmark, 2000a, 2000b). Hypothesized to result from changing  
40 ionization of the atmosphere from solar-modulated cosmic ray fluxes, an empirical association of cloud  
41 cover variations during 1984–1990 and the solar cycle remains controversial because of uncertainties about  
42 the reality of the decadal signal itself, the phasing or anti-phasing with solar activity, and its separate  
43 dependence for low, mid and high clouds. In particular, the cosmic ray time series does not correspond to  
44 global total cloud cover after 1991 or to global low-level cloud cover after 1994 (Kristjánsson and  
45 Kristiansen, 2000; Sun and Bradley, 2002) without unproven detrending (Usoskin et al., 2004). Furthermore,  
46 the correlation is significant with low-level cloud cover based only on infrared (not visible) detection. Nor do  
47 multidecadal (1952–1997) time series of cloud cover from ship synoptic reports exhibit a relationship to  
48 cosmic ray flux. However, there appears to be a small but statistically significant positive correlation  
49 between cloud over the UK and galactic cosmic ray flux during 1951–2000 (Harrison and Stephenson,  
50 2006). Contrarily, cloud cover anomalies from 1900–1987 over the United States do have a signal at 11-  
51 years that is anti-phased with the galactic cosmic ray flux (Udelhofen and Cess, 2001). Because the  
52 mechanisms are uncertain, the apparent relationship between solar variability and cloud cover has been  
53 interpreted to result not only from changing cosmic ray fluxes modulated by solar activity in the heliosphere  
54 (Usoskin et al., 2004) and solar induced changes in ozone (Udelhofen and Cess, 2001), but also as a result of  
55 sea surface temperatures altered directly by changing total solar irradiance (Kristjánsson et al., 2002) and  
56 internal variability by El Niño Southern Oscillation (Kerthaler et al., 1999). In reality, different direct and  
57 indirect physical processes (such as those described in Chapter 9, Section 9.2) may operate simultaneously.

1  
2 The direct RF due to increase in solar irradiance is reduced from the TAR. The best estimate is  $+0.12 \text{ W m}^{-2}$   
3 (90% confidence interval:  $0.06$  to  $0.30 \text{ W m}^{-2}$ ). While there have been advancements in the direct solar  
4 irradiance variation, there remain large uncertainties. The level of scientific understanding is elevated to *low*  
5 relative to TAR for solar forcing due to direct irradiance change, while declared as *very low* for cosmic ray  
6 influences (Table 2.11).  
7

## 8 **2.7.2 Explosive Volcanic Activity**

### 9 **2.7.2.1 Radiative Effects of Volcanic Aerosols**

10  
11  
12 Volcanic sulphate aerosols are formed as a result of oxidation of the sulphur gases emitted by explosive  
13 volcanic eruptions into the stratosphere. The process of gas-to-particle conversion has an e-folding time of  
14 roughly 35 days (Bluth et al., 1992; Read et al., 1993). The e-folding time (by mass) for sedimentation of  
15 sulfate aerosols is typically about 12 to 14 months (Lambert et al., 1993; Baran and Foot, 1994; Bluth et al.,  
16 1997; Barnes and Hoffman, 1997). Also emitted directly during an eruption are volcanic ash particulates  
17 (siliceous material). These are particles usually larger than  $2 \mu\text{m}$  that sediment out of the stratosphere fairly  
18 rapidly due to gravity (within 3 months or so), but could also play a role in the radiative perturbations in the  
19 immediate aftermath of an eruption. Stratospheric aerosol data incorporated for climate change simulations  
20 tends to be mostly that of the sulphates (Sato et al., 1993; Stenchikov et al., 1998; Ramachandran et al.,  
21 2000; Hansen et al., 2002; Tett et al., 2002; Ammann et al., 2003). As noted in the SAR and the TAR,  
22 explosive volcanic events are episodic, but the stratospheric aerosols resulting from them yield substantial  
23 transitory perturbations to the radiative energy balance of the planet, with both shortwave and longwave  
24 effects sensitive to the microphysical characteristics of the aerosols (e.g., size distribution).  
25

26 Long-term ground-based and balloonborne instrumental observations have resulted in an understanding of  
27 the optical effects and microphysical evolution of volcanic aerosols (Deshler et al., 2003; Hofmann et al.,  
28 2003). Important ground-based observations of aerosol characteristics from pre-satellite era spectral  
29 extinction measurements have been analyzed by Stothers (2001a,b), but they do not provide a global  
30 coverage. Global observations of stratospheric aerosol over the last 25 years have been possible owing to a  
31 number of satellite platforms e.g., TOMS and TOVS have been used to estimate  $\text{SO}_2$  loadings from volcanic  
32 eruptions (Krueger et al., 2000; Prata et al., 2003). The SAGE and SAM projects (e.g., McCormick, 1987)  
33 have provided vertically resolved stratospheric aerosol spectral extinction data for over 20 years, the longest  
34 such record. This dataset has a significant gap in coverage at the time of the El Chichón eruption in 1982 (the  
35 second most important in the 20th century after Mt. Pinatubo in 1991), and when the aerosol cloud is dense;  
36 these gaps have been partially filled by lidar measurements and field campaigns (e.g., Antuña et al., 2003;  
37 Thomason and Peter, 2006).  
38

39 Volcanic aerosols transported in the atmosphere to polar regions are preserved in the ice sheets, thus  
40 recording the history of the Earth's volcanism for thousands of years (Bigler et al., 2002; Palmer et al., 2002;  
41 Mosley-Thompson et al., 2003). However, the atmospheric loadings obtained from ice records suffer from  
42 uncertainties due to: imprecise knowledge of the latitudinal distribution of the aerosols; depositional noise  
43 which can affect the signal for an individual eruption in a single ice core; and poor constraints on aerosol  
44 microphysical properties.  
45

46 The best documented explosive volcanic event to-date, by way of reliable and accurate observations, is the  
47 1991 eruption of Mt. Pinatubo. The growth and decay of aerosols resulting from this eruption have provided  
48 a basis for modelling the RF due to explosive volcanoes. There have been no explosive and climatically-  
49 significant volcanic events since Pinatubo. As pointed out in Ramaswamy et al. (2001), stratospheric aerosol  
50 concentrations are now at the lowest concentrations since the satellite era and global coverage began in about  
51 1980. Altitude-dependent stratospheric optical observations at a few wavelengths, together with columnar  
52 optical and physical measurements, have been used to construct the time-dependent global field of  
53 stratospheric aerosol size distribution formed in the aftermath of volcanic events. The wavelength-dependent  
54 stratospheric aerosol single-scattering characteristics calculated for the solar and longwave spectrum are  
55 deployed in the climate models to account for the resulting radiative (shortwave plus longwave)  
56 perturbations.  
57

1 Using available satellite and ground based observations; Hansen et al. (2002) have constructed a volcanic  
2 aerosols data set for the 1850–1999 period (Sato et al., 1993). This has yielded zonal-mean vertically  
3 resolved aerosol optical depths for visible wavelengths and column-average effective radii. Stenchikov et al.  
4 (2006) introduced a slight variation to this dataset employing UARS observations to modify the effective  
5 radii relative to Hansen et al. (2002), thus accounting for variations with altitude. Ammann et al. (2003) have  
6 developed a dataset of total aerosol optical depth for the period since 1890 that does not include the Krakatau  
7 eruption. This is based on empirical estimates of atmospheric loadings which are then globally distributed  
8 using a simplified parameterization of atmospheric transport, and employ a fixed aerosol effective radius  
9 (0.42  $\mu\text{m}$ ) for calculating optical properties. The above datasets have essentially provided the bases for the  
10 volcanic aerosols implemented in virtually all of the models that have performed the 20th century climate  
11 integrations (Stenchikov et al., 2006). Relative to Sato et al. (1993), the Ammann et al. (2003) estimate  
12 yields a larger value of the optical depth, by 20 to 30% in the second part of the 20th century, and by 50%  
13 for eruptions at the end of 19th and beginning of 20th century e.g., Santa Maria (1902) (Figure 2.18).

14  
15 [INSERT FIGURE 2.18 HERE]

16  
17 The global-mean RF calculated using the Sato et al. (1993) data yields a peak in radiative perturbation of  
18 about  $-3 \text{ W m}^{-2}$  for the strong (rated in terms of emitted  $\text{SO}_2$ ) 1860 and 1991 eruptions of Krakatau and  
19 Pinatubo, respectively. The value is reduced to about  $-2 \text{ W m}^{-2}$  for the relatively less intense El Chichón and  
20 Agung eruptions (Hansen et al., 2002). As expected from the arguments above, Ammann's RF is roughly 20–  
21 30% larger than Sato's RF.

22  
23 Not all features of the aerosols are well quantified and extending and improving the data sets remains an  
24 important area of research. This includes improved estimates of the aerosol size parameters (Bingen et al.,  
25 2004), a new approach for calculating aerosol optical characteristics using SAGE and UARS data (Bauman  
26 et al., 2003), and intercomparison of data from different satellites and combining them to fill gaps (Randall et  
27 al., 2001). While the aerosol characteristics are better constrained for the Pinatubo eruption, and to some  
28 extent for El Chichón and Agung eruptions, the reliability degrades for aerosols from explosive volcanic  
29 events further back in time as there are little, if any, observational constraints on their optical depth and size  
30 evolution.

31  
32 The radiative effects due to volcanic aerosols from major eruptions are manifest in the global mean anomaly  
33 of reflected solar radiation; this variable affords a good estimate of radiative effects that can actually be  
34 tested against observations. However, unlike RF, this variable contains effects due to feedbacks (e.g.,  
35 changes in cloud distributions) so that it is actually more a signature of the climate response. In the case of  
36 the Pinatubo eruption, with a peak global visible optical depth of about 0.15, simulations yield a large  
37 negative perturbation as noted above of about  $-3 \text{ W m}^{-2}$  (Ramachandran et al., 2000; Hansen et al., 2002)  
38 (see also Chapter 9, Section 9.2). This modelled estimate of reflected solar radiation compares reasonably  
39 with ERBS observations (Minnis et al., 1993). However, the ERBS observations were for a relatively short  
40 duration, and the model-observation comparisons are likely affected by differing cloud effects in simulations  
41 and measurements. It is interesting to note (Stenchikov et al., 2006) that, in the Pinatubo case, the GISS  
42 models that use the Sato et al. (1993) data yield an even greater solar reflection than the NCAR model which  
43 uses the larger (Ammann et al., 2003) optical depth estimate.

#### 44 45 2.7.2.2 *Thermal, Dynamical and Chemistry Perturbations Forced by Volcanic Aerosols*

46  
47 Four distinct mechanisms have been invoked with regards to the climate response to volcanic aerosol RF.  
48 First, these forcings can directly affect the Earth's radiative balance and thus alter surface temperature.  
49 Second, they introduce horizontal and vertical heating gradients; these can alter the stratospheric circulation,  
50 in turn affecting the troposphere. Third, the forcings can interact with internal climate system variability  
51 (e.g., El Niño Southern Oscillation, North Atlantic Oscillation, Quasi Biennial Oscillation) and dynamical  
52 noise, thereby triggering, amplifying or shifting these modes (see Chapter 9, Section 9.2; Yang and  
53 Schlesinger, 2001; Stenchikov et al., 2004). Fourth, volcanic aerosols provide surfaces for heterogeneous  
54 chemistry affecting global stratospheric ozone distributions (Chipperfield et al., 2003) and perturbing other  
55 trace gases for a considerable period following an eruption. Each of the above mechanisms has its own  
56 spatial and temporal response pattern. Also, the mechanisms could depend on the background state of the

1 climate system, and thus on other forcings (e.g., due to well-mixed gases, Meehl et al., 2004), or interact  
2 with each other.

3  
4 The complexity of radiative-dynamical response forced by volcanic impacts suggests that it is important to  
5 calculate aerosol radiative effect interactively within the model rather than prescribe them (Andronova et al.,  
6 1999; Broccoli et al., 2003). Despite differences in volcanic aerosol parameters employed, models  
7 computing the aerosol radiative effects interactively yield tropical and global-mean lower stratospheric  
8 warming that are fairly consistent with each other and with observations (Yang and Schlesinger, 2002;  
9 Stenchikov et al., 2004; Ramachandran et al., 2000; Hansen et al., 2002; Ramaswamy et al., 2006a);  
10 however, there is a considerable range in the responses in the polar stratosphere and troposphere. The global-  
11 mean warming of the lower stratosphere is due mainly to aerosol effects in the longwave spectrum, in  
12 contrast to the flux changes at the top-of-the-atmosphere that are essentially due to aerosol effects in the  
13 solar spectrum. The net radiative effects of volcanic aerosols on the thermal and hydrologic balance (e.g.,  
14 surface temperature and moisture) has been highlighted by recent studies (Free and Angell, 2002; Jones et  
15 al., 2003; see Chapter 6; see Chapter 9 for significance of the simulated responses, and model-observation  
16 comparisons for 20th century eruptions). A mechanism closely linked to the optical depth perturbation and  
17 ensuing warming of the tropical lower stratosphere is the potential change in the cross-tropopause water  
18 vapour flux (Joshi and Shine, 2003, see Section 2.3.7).

19  
20 Anomalies in the volcanic aerosol-induced global radiative heating distribution can force significant changes  
21 in atmospheric circulation e.g., perturbing the equator-to-pole heating gradient (Stenchikov et al., 2002;  
22 Ramaswamy et al., 2006b; see Chapter 9, Section 9.2) and forcing a positive phase of the Arctic Oscillation  
23 that in turn causes a counterintuitive boreal winter warming in middle and high latitudes over Eurasia and  
24 North America (Perlwitz and Graf, 2001; Stenchikov et al., 2002, 2004, 2006; Shindell et al., 2003b, 2004;  
25 Perlwitz and Harnik, 2003; Rind et al., 2005; Miller et al., 2006).

26  
27 Stratospheric aerosols affect the chemistry and transport processes in the stratosphere, resulting in the  
28 depletion of ozone (Brasseur and Granier, 1992; Tie et al., 1994; Solomon et al., 1996; Chipperfield et al.,  
29 2003). Stenchikov et al. (2002) demonstrate a link between ozone depletion and Arctic Oscillation response;  
30 this is essentially a secondary radiative mechanism induced by volcanic aerosols through stratospheric  
31 chemistry. Stratospheric cooling in the polar region associated with a stronger polar vortex initiated by  
32 volcanic effects can increase the probability of formation of polar stratospheric clouds and therefore enhance  
33 the rate of heterogeneous chemical destruction of stratospheric ozone, especially in the northern hemisphere  
34 (Tabazadeh et al., 2002). The above studies indicate effects on the stratospheric ozone layer in the wake of a  
35 volcanic eruption and under conditions of enhanced anthropogenic halogen loading. Interactive  
36 microphysics-chemistry-climate models (Rozanov et al., 2002, 2004; Timmreck et al., 2003; Shindell et al.,  
37 2003b; Dameris et al., 2005) indicate that aerosol-induced stratospheric heating affects the dispersion of the  
38 cloud, thus affecting the spatial RF. However the models' simplified treatment of aerosol microphysics  
39 introduces biases; further, they usually overestimate the mixing at the tropopause level and intensity of  
40 meridional transport in the stratosphere (Schoeberl et al., 2003; Douglass et al., 2003). For present climate  
41 studies, it is practical to utilize simpler approaches that are reliably constrained by aerosol observations.

42  
43 Because of its episodic and transitory nature, it is difficult to give a best estimate for the volcanic RF, unlike  
44 the other agents. Neither a best estimate nor a level of scientific understanding rank was given in the TAR.  
45 For the well documented case of the explosive 1991 Mt. Pinatubo eruption, there is a good scientific  
46 understanding. However, the limited knowledge of the RF associated with prior episodic, explosive events  
47 indicates a *low* level of scientific understanding (Table 2.11).

## 48 49 **2.8 Utility of Radiative Forcing**

50  
51 The TAR and other assessments have concluded that RF is a useful tool for estimating, to a first order, the  
52 relative global climate impacts of differing climate-change mechanisms (Ramaswamy et al., 2001; Jacob et  
53 al., 2005). In particular RF can be used to estimate the relative equilibrium globally averaged surface  
54 temperature change due to different forcing agents. However, RF is not a measure of other aspects of climate  
55 change or the role of emissions (see Sections 2.2 and 2.10). Previous GCM studies have indicated that the  
56 climate sensitivity parameter was more or less constant (varying by less than 25%) between mechanisms  
57 (Ramaswamy et al., 2001; Chipperfield et al., 2003). However, this level of agreement was found not to hold

1 for certain mechanisms such as ozone changes at some altitudes and changes in absorbing aerosol. Because  
2 the climate responses, and in particular the equilibrium climate sensitivities, exhibited by GCMs, vary by  
3 much more than 25% (see Chapter 9, Section 9.6), Ramaswamy et al. (2001) and Jacob et al. (2005)  
4 concluded that RF is the most simple and straightforward measure for the quantitative assessment of climate  
5 change mechanisms, especially for the LLGHGs. This section discusses the several studies since the TAR  
6 that have examined the relationship between RF and climate response. Note that this assessment is entirely  
7 based on climate model simulations.

### 8 9 **2.8.1 Vertical Forcing Patterns and Surface Energy Balance Changes**

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

### 23 24 25 **2.8.2 Spatial Patterns of Radiative Forcing**

26 Each RF agent has a unique spatial pattern (see Ramaswamy et al., 2001, Figure 6.7 for examples). When  
27 combining RF agents it not just the global-mean RF that needs to be considered. For example, even with a  
28 net global mean RF of zero, significant regional RFs can be present and these can affect the global mean  
29 temperature response (see Section 2.8.5). RF spatial patterns also affect the pattern of climate response.  
30 However, note also that, to first order, very different RF patterns can have similar patterns of surface  
31 temperature response and the location of maximum RF is rarely coincident with the location of maximum  
32 response (Boer and Yu., 2003b). Identification of different patterns of response is particularly important for  
33 attributing past climate change to particular mechanisms, and is also important for the prediction of regional  
34 patterns of future climate change. This chapter employs RF as the method for ranking the effect of a forcing  
35 agent on the equilibrium global temperature change, and we only this aspect of the forcing-response  
36 relationship is discussed. However, patterns of RF are presented as a diagnostic in Section 2.9.5.

### 37 38 39 **2.8.3 Alternative Methods of Calculating Radiative Forcing**

40 RFs are increasingly being diagnosed from GCM integrations where the calculations are complex (Stuber et  
41 al., 2001a; Tett et al., 2002; Gregory et al., 2004). This chapter also discusses several mechanisms that  
42 include some response in the troposphere, such as cloud changes. These mechanisms are not initially  
43 radiative in nature, but will eventually lead to a radiative perturbation of the surface-troposphere system, that  
44 could conceivably be measured at the top of the atmosphere. Jacob et al. (2005) refer to these mechanisms as  
45 non-radiative forcings (see also Section 2.2). Alternatives to the standard stratospherically-adjusted RF  
46 definition have been proposed that may help account for these processes. Since the TAR, several studies  
47 have employed GCMs to diagnose the zero-surface-temperature-change RF (see Figure 2.2 and Section 2.2).  
48 These studies have used a number of different methodologies. Shine et al. (2003) fixed both land and sea  
49 surface temperatures globally and calculated a radiative energy imbalance: this technique is only feasible in  
50 GCMs with relatively simple land-surface parameterizations. Hansen et al. (2005) fixed sea-surface  
51 temperatures and calculated an RF by adding an extra term to the radiative imbalance that takes into account  
52 how much the land surface temperatures had responded. Sokolov (2006) diagnosed this RF by computing  
53 surface-only and atmospheric-only components of climate feedback separately in a slab model and then  
54 modifying the stratospherically adjusted RF by the atmospheric-only feedback component. Gregory et al.  
55 (2004) (see also Hansen et al., 2005 and Forster and Taylor, 2006) used a regression method with a globally-  
56 averaged-temperature-change ordinate to diagnose this RF: this method had the largest uncertainties. Shine  
57

1 et al. (2003), Hansen et al. (2005) and Sokolov (2006) all find that that the fixed-surface-temperature RF is a  
2 better predictor of the equilibrium global mean surface temperature response than the stratospherically  
3 adjusted RF. Further, it was a particularly useful diagnostic for changes in absorbing aerosol where the  
4 stratospherically adjusted RF could fail as a predictor of the surface temperature response (see Section  
5 2.8.5.5). Differences between the zero-surface-temperature-change RF and the stratospheric-adjusted RF can  
6 be caused by semi-direct and cloud-aerosol interaction effects beyond the cloud-albedo RF. For most  
7 mechanisms, aside from the case of certain aerosol changes, the difference is *likely* to be small (Shine et al.,  
8 2003; Hansen et al., 2005; Sokolov, 2006). These calculations also remove problems associated with  
9 defining the tropopause in the stratospherically-adjusted RF definition (Shine et al., 2003; Hansen et al.,  
10 2005). However, stratospheric-adjusted RF has the advantage that it does not depend on relatively uncertain  
11 components of a GCMs response, such as cloud changes. For the LLGHGs the stratospheric-adjusted RF  
12 also has the advantage that it is also readily calculated in detailed off-line radiation codes. For these reasons  
13 the stratospherically adjusted RF is retained as the measure of comparison used in this chapter (see Section  
14 2.2). However, to first order all methods are comparable and all prove useful for understanding climate  
15 response.

#### 17 2.8.4 *Linearity of the Forcing-Response Relationship*

18  
19 Reporting findings from several studies, the TAR concluded that responses to individual RFs could be  
20 linearly added to gauge the global mean response, but not necessarily the regional response (Ramaswamy et  
21 al., 2001). Since then studies with several equilibrium and/or transient integrations of several different  
22 GCMs have found no evidence of any non-linearity for changes in greenhouse gas and sulphate aerosol  
23 (Boer and Yu, 2003b; Gillett et al., 2004; Matthews et al., 2004; Meehl et al., 2004). Two of these studies  
24 also examined realistic changes in many other forcing agents without finding evidence of a non-linear  
25 response (Meehl et al., 2004; Matthews et al., 2004). In all four studies, even the regional changes typically  
26 added linearly. However, Meehl et al. (2004) observed that neither precipitation changes nor all regional  
27 temperature changes were linearly additive. This linear relationship also breaks down for global mean  
28 temperatures when aerosol-cloud interactions beyond the cloud-albedo RF are included in GCMs (Feichter et  
29 al., 2004; see also Rotstayn and Penner, 2001 and Lohmann and Feichter, 2005). Studies which include these  
30 effects modify clouds in their models, producing an additional radiative imbalance. Rotstayn and Penner  
31 (2001) find that if these aerosol-cloud effects are accounted for as additional forcing terms, the inference of  
32 linearity can be restored (see Sections 2.8.3 and 2.8.5). Studies also find non linearities for large negative  
33 RFs, where static stability changes in the upper troposphere affect the climate feedback (e.g., Hansen et al.,  
34 2005). For the magnitude and range of realistic RFs discussed in this chapter, and excluding cloud-aerosol  
35 interaction effects, there is high confidence of a linear relationship between global mean RF and global mean  
36 surface temperature response.

#### 38 2.8.5 *Efficacy and Effective Radiative Forcing*

39  
40 Efficacy (E) is defined as the ratio of the climate sensitivity parameter for a given forcing agent ( $\lambda_i$ ) to the  
41 climate sensitivity parameter for CO<sub>2</sub> changes, i.e.  $E_i = \lambda_i / \lambda_{CO_2}$  (Joshi et al., 2003; Hansen and Nazarenko,  
42 2004). Efficacy can then be used to define an effective RF ( $=E_i RF_i$ ) (Joshi et al., 2003; Hansen et al. 2005).  
43 For the effective RF, the climate sensitivity parameter is independent of the mechanism, so comparing this  
44 forcing is equivalent to comparing the equilibrium global mean surface temperature change. That is,  $\Delta T_s =$   
45  $\lambda_{CO_2}(E_i RF_i)$ . Preliminary studies have found that efficacy values for a number of forcing agents show less  
46 model dependency than the climate sensitivity values (Joshi et al., 2003). Effective RFs have been used get  
47 one step closer to an estimator of the likely surface temperature response than can be achieved by using RF  
48 alone (Sausen and Schumann, 2000; Hansen et al., 2005; Lohmann and Feichter, 2005). Adopting the zero-  
49 surface-temperature change RF, which has efficacies closer to unity, may be another way of achieving  
50 similar goals (see Section 2.8.3). This section assesses the efficacy associated with stratospherically-adjusted  
51 RF, as this is the definition of RF adopted in this chapter (see Section 2.2). Therefore, cloud aerosol  
52 interaction effects beyond the cloud albedo RF are included in the efficacy term. The findings presented in  
53 this section are from an assessment of all the studies referenced in the caption of Figure 2.19, which presents  
54 a synthesis of efficacy results. As space is limited not all these studies are explicitly discussed in the main  
55 text.

56  
57 [INSERT FIGURE 2.19 HERE]

### 2.8.5.1 *Generic Understanding*

Since the TAR several 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 studies 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, 2006; Stuber et al., 2005). Many of the studies presented in Figure 2.19 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, 2006; Stuber et al., 2005). Nearly all studies that examine it find that high latitude forcings have higher efficacies than tropical forcings. 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, 2006; 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, 2006).

### 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 2.19). Further, the most recent result with the NCAR CCM3 GCM (Govindasamy et al., 2001b) indicates 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., 2005) which are slightly smaller than one. Overall there is medium 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<sub>2</sub>, have less high latitude RF, and with more of the RF 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 RF find efficacies of 1.0 or smaller. One study finds a smaller efficacy than other models (0.63: Gregory et al., 2004). However, their unique methodology for calculating climate sensitivity has large uncertainties, (see Section 2.8.4). These studies have only examined solar RF from a total solar irradiance change; any indirect solar effects (see Section 2.7.1.3) are not included in this efficacy estimate. Overall there is medium confidence that the direct solar efficacy is within the 0.7–1.0 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 such 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. Only one study has used realistic stratospheric ozone changes (see Figure 2.19); thus, our knowledge is still incomplete. Conclusions are only drawn from the idealised studies where there is (1) medium confidence that the efficacy is within a 0.5–2.0 range and (2) established but incomplete physical understanding of how and why the efficacy could be larger than 1.0. There is medium confidence that for realistic tropospheric ozone perturbations the efficacy is within the 0.6–1.1 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 there is medium confidence that efficacies for scattering aerosol would be in the 0.7–1.1 range. Efficacies are *likely* to be similar for scattering aerosol in the troposphere and stratosphere.

With the formulation of RF employed in this chapter, the efficacy of the cloud-albedo RF accounts for cloud lifetime effects (Section 2.8.3). Only two studies contained enough information to calculate efficacy in this way and both find efficacies higher than 1.0. However, the uncertainties in quantifying the cloud lifetime effect make this efficacy very uncertain. If cloud-lifetime effects were excluded from the efficacy term, the cloud albedo efficacy would *very likely* be similar to that of the direct effect (see Figure 2.19).

### 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., 2006b). 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.; 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 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) (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 dependent 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, 2006). Further Penner et al. (2006b) 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 stratospheric-adjusted definition of RF (see Section 2.8.3).

### 2.8.5.7 *Other Forcing Agents*

Efficacies for some other effects have been evaluated by one or two modelling groups. Hansen et al. (2005) found that land-use albedo RF had an efficacy of roughly 1.0, whilst the BC-snow albedo RF had an efficacy of 1.7. Ponater et al. (2005) find an efficacy of 0.6 for contrail RF and this agrees with a suggestion from Hansen et al. (2005) that high cloud changes should have smaller efficacies. 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*

Efficacy is a new concept introduced since the TAR; its physical understanding is becoming established (see Section 2.8.5). When employing the stratospherically-adjusted RF, there is medium confidence that efficacies are within the 0.75–1.25 range for most realistic RF mechanisms aside from aerosol and

1 stratospheric ozone changes. There is medium confidence that realistic aerosol and ozone changes have  
2 efficacies within the 0.5–2.0 range. Further, zero-surface temperature change RFs are *very likely* to have  
3 efficacies significantly closer to 1.0 for all mechanisms. It should be noted that efficacies have only been  
4 evaluated in GCMs and actual climate efficacies could be different than those quoted in Section 2.8.5.

## 6 2.9 Synthesis

7  
8 This section begins by synthesizing the discussion of the RF concept. It presents summaries of the global  
9 mean RFs assessed in earlier sections and discusses time evolution and spatial patterns of RF. It also presents  
10 a brief synthesis of *surface forcing* diagnostics. It breaks down the analysis of RF several ways to aid and  
11 advance the understanding of the drivers of climate change.

12  
13 RFs are calculated in various ways depending on the agent: from changes in emissions and/or changes in  
14 concentrations; and from observations and other knowledge of climate change drivers. Current RF depends  
15 on present day concentrations of a forcing agent, which in turn depends on the past history of emissions.  
16 Some climate response to these RFs can be expected to have already occurred. Additionally, as RF is a  
17 comparative measure of equilibrium climate change and the Earth's climate is not in an equilibrium state,  
18 additional climate change in the future is also expected from present day RFs (see Section 2.2 and Chapter  
19 10, Section 10.7). As previously stated in Section 2.2, RF alone is not a suitable metric for weighting  
20 emissions, for this purpose the lifetime of the forcing agent also needs to be considered (see Sections 2.9.4  
21 and 2.10).

22  
23 RFs are considered external to the climate system (see Section 2.2). Aside from the natural RFs (solar,  
24 volcanoes), the other RFs are considered to be anthropogenic (i.e., directly attributable to human activities).  
25 For the LLGHGs it is assumed that all changes in their concentrations since preindustrial times are human-  
26 induced (either directly through emissions or from land-use changes); these concentration changes are used  
27 to calculate the RF. Likewise stratospheric ozone changes are also taken from satellite observations and  
28 changes are primarily attributed to Montreal Protocol controlled gases, although there may also be a climate  
29 feedback contribution to these trends (see Section 2.3.4). For the other RFs, anthropogenic emissions and/or  
30 human-induced land-use changes are used in conjunction with Chemical Transport Models and/or GCMs to  
31 estimate the anthropogenic RF.

### 33 2.9.1 Uncertainties in Radiative Forcing

34  
35 The TAR assessed uncertainties in global-mean RF by attaching an error bar to each RF term that was  
36 “*guided by the range of published values and physical understanding*”. It also quoted a level of scientific  
37 understanding (LOSU) for each RF, which was a subjective judgment of the estimate's reliability.

38  
39 The concept of LOSU has been slightly modified based on the IPCC AR4 uncertainty guidelines. Error bars  
40 now represent the 5–95% (90%) confidence range (see Box TS-1 on uncertainty in the Technical Summary).  
41 Only “well established” RFs are quantified. “Well established” implies that there is qualitatively both  
42 sufficient evidence and sufficient consensus from published results to estimate a central RF estimate and a  
43 range. “Evidence” is assessed by an A-C grade, with an A grade implying strong evidence and C insufficient  
44 evidence. Strong evidence implies that observations have verified aspects of the RF mechanism and that  
45 there is a sound physical model to explain the RF. “Consensus” is assessed by assigning a number between  
46 1–3, where 1 implies a good deal of consensus and 3 insufficient consensus. This ranks the number of studies  
47 and how well studies agree on quantifying the RF, and especially how well observation-based studies agree  
48 with models. The product of “Evidence” and “Consensus” factors give the LOSU rank. These ranks are *high*,  
49 *medium*, *medium-low*, *low* or *very-low*. Ranks of *very-low* are not evaluated. The quoted 90% confidence  
50 range of RF quantifies the value uncertainty, as derived from the expert assessment of published values and  
51 their ranges. For most RFs, many studies have now been published, which generally makes the sampling of  
52 parameter space more complete and the value uncertainty more realistic, compared to the TAR. This is  
53 particularly true for both the direct and cloud-albedo aerosol RF (see Section 2.4). Table 2.11 summarises  
54 the key certainties and uncertainties and indicates the basis for the 90% confidence range estimate. Note that  
55 the aerosol terms will have added uncertainties due to the uncertain semi-direct and cloud-lifetime effects.  
56 These uncertainties in the response to the RF (efficacy) are discussed in Section 2.8.5.

1 Table 2.11 indicates that there is now stronger evidence for most of the RFs being discussed in this chapter.  
2 Some effects are not quantified, either because they do not have enough evidence or their quantification  
3 lacks consensus. These include certain mechanisms associated with land-use, stratospheric water vapour and  
4 cosmic rays. Cloud-lifetime and the semi-direct effects are also excluded from this analysis as they are  
5 deemed to be part of the climate response (see Chapter 7, Section 7.5). The RFs from the LLGHGs have both  
6 a high degree of consensus and a very large amount of evidence and, thereby, place our understanding of  
7 these effects at a considerably higher level than any other effect.

8  
9 [INSERT TABLE 2.11 HERE]

## 10 2.9.2 Global Mean Radiative Forcing

11 The RFs discussed in this chapter, their uncertainty ranges, and efficacies are summarized in Figure 2.20 and  
12 Table 2.12. RFs from forcing agents have been combined into their main groupings. This is particularly  
13 useful for aerosol as its total direct RF is considerably better constrained than the RF from individual aerosol  
14 types (see Section 2.4.4). Table 2.1 gives a further component breakdown of RF for the LLGHGs. RFs are  
15 the stratospherically-adjusted RF and they have not been multiplied by efficacies (see Sections 2.2 and 2.8).

16  
17 [INSERT FIGURE 2.20 HERE]

18  
19 [INSERT TABLE 2.12 HERE]

20  
21 In the TAR no estimate of the total combined RF from all anthropogenic forcing agents was given because:  
22 a) some of the forcing agents did not have central or best estimate; b) a degree of subjectivity was included  
23 in the error estimates; c) uncertainties associated with the linear additivity assumption and efficacy had not  
24 been evaluated. Some of these limitations still apply. However, methods for objectively adding the RF of  
25 individual species have been developed (e.g. Schwartz and Andreae, 1996; Boucher and Haywood, 2001).  
26 Also, as efficacies are now better understood and quantified (see Section 2.8.5), and as the linear-additivity  
27 assumption has been more thoroughly tested (see Section 2.8.4), it becomes scientifically justifiable for RFs  
28 from different mechanisms to be combined, with certain exceptions as noted below. Adding together the  
29 anthropogenic RF values shown in panel a) of Figure 2.20 and combining their individual uncertainties,  
30 gives the probability density functions (PDFs) of RF that are shown in panel b). Three PDFs are shown: the  
31 combined RF from greenhouse gas changes (LLGHGs and ozone); the combined direct aerosol and cloud  
32 albedo RFs and the combination of all anthropogenic RFs. The solar RF is not included in any of these  
33 distributions. The PDFs are generated by combining the 90% confidence estimates for the RFs, assuming  
34 independence and employing a one million point Monte-Carlo simulation to derive the PDFs (see Boucher  
35 and Haywood, 2001, and Figure 2.20 caption for details).

36  
37 The PDFs show that LLGHGs and ozone contribute a positive RF of  $2.9 \pm 0.3 \text{ W m}^{-2}$ . The combined aerosol  
38 direct and cloud-albedo effect exert an RF that is *virtually certain* to be negative, with a median RF of  $-1.3$   
39  $\text{W m}^{-2}$  and a  $-2.2$  to  $-0.5 \text{ W m}^{-2}$  90% confidence range. The asymmetry in the combined aerosol PDF is  
40 caused by the estimates in Tables 2.6 and 2.7 being non-Gaussian. The combined net RF estimate for all  
41 anthropogenic drivers has a value of  $1.6 \text{ W m}^{-2}$  with a  $0.6$  to  $2.4 \text{ W m}^{-2}$  90% confidence range. Note that the  
42 RFs from surface albedo change, stratospheric water vapour change and persistent contrails are only  
43 included in the combined anthropogenic PDF and not the other two.

44  
45 Statistically, the PDF shown in Figure 2.20 indicates just a 0.2% probability that the total RF from  
46 anthropogenic agents is negative, which would suggest that it is *virtually certain* that the combined RF from  
47 anthropogenic agents is positive. Additionally, the PDF presented here suggests that it is *extremely likely* that  
48 the total anthropogenic RF is larger than  $0.6 \text{ W m}^{-2}$ . This combined anthropogenic PDF is better constrained  
49 than that shown in Boucher and Haywood (2001) because each of the individual RFs have been quantified to  
50 90% confidence levels enabling a more definite assessment, and because the uncertainty in some of the RF  
51 estimates is considerably reduced. For example, modelling of the total direct RF due to aerosols is better  
52 constrained by satellite and surface based observations (Section 2.4.2), and the current estimate of the cloud-  
53 albedo indirect effect has a best estimate and uncertainty associated with it, rather than just a range. The  
54 LLGHG RF has also increased by  $0.20 \text{ W m}^{-2}$  since 1998, making a positive RF more likely than in Boucher  
55 and Haywood (2001).

1  
2 Nevertheless, there are some structural uncertainties associated with the assumptions used in the construction  
3 of the PDF and the assumptions describing the component uncertainties. Normal distributions are assumed  
4 for most RF mechanisms (with the exceptions noted in the caption); this may not accurately capture  
5 extremes. Additionally, as in Boucher and Haywood (2001), all of the individual RF mechanisms are given  
6 equal weighting, even though the level of scientific understanding differs between forcing mechanisms. Note  
7 also that variation in efficacy and hence the semi-direct and cloud-lifetime effects are not accounted for, as  
8 these are not considered to be RFs in this report (see Section 2.2). Adding these effects, together with other  
9 potential mechanisms that have so far not been defined as RFs and quantified, would introduce further  
10 uncertainties but give a fuller picture of the role of anthropogenic drivers. Introducing efficacy would give a  
11 broader PDF and a large cloud-lifetime effect would reduce the median estimate. Despite these caveats, from  
12 the current knowledge of individual forcing mechanisms presented here it remains *extremely likely* that the  
13 combined anthropogenic RF is both *positive* and *substantial* (best-estimate:  $+1.6 \text{ W m}^{-2}$ ).  
14

### 15 **2.9.3 Global-Mean Radiative Forcing by Emission Precursor**

16  
17 The RF due to changes in the concentration of a single forcing agent can have contributions from emissions  
18 of several compounds (Shindell et al., 2005). The RF of methane, for example, is affected by methane  
19 emissions, as well as  $\text{NO}_x$  emissions. The methane RF quoted in Table 2.12 and shown in Figure 2.20 is a  
20 value that combines the effects of both emissions. As an anthropogenic or natural emission can affect several  
21 forcing agents, it is useful to assess the current RF caused by each primary emission. For example, emission  
22 of  $\text{NO}_x$  affects methane, tropospheric ozone, and tropospheric aerosols. Based on a development, carried  
23 forward from the TAR, this section assesses the RF terms associated with each principal emission including  
24 indirect RFs related to perturbations of other forcing agents, with the results shown in Figure 2.21. The  
25 following indirect forcing mechanisms are considered.  
26

- 27 • Fossil carbon from non- $\text{CO}_2$  gaseous compounds, which eventually increase  $\text{CO}_2$  in the atmosphere  
28 (from  $\text{CO}$ ,  $\text{CH}_4$ , and NMVOCs emissions)
- 29 • Changes in stratospheric ozone (from  $\text{N}_2\text{O}$  and halocarbon (CFCs, HCFC, halons, etc.) emissions)
- 30 • Changes in tropospheric ozone (from  $\text{CH}_4$ ,  $\text{NO}_x$ ,  $\text{CO}$ , and NMVOCs emissions)
- 31 • Changes in OH affecting the lifetime of  $\text{CH}_4$  (from  $\text{CH}_4$ ,  $\text{CO}$ ,  $\text{NO}_x$ , and NMVOCs emissions)
- 32 • Changing nitrate and sulphate aerosols through changes in  $\text{NO}_x$  and  $\text{SO}_2$  emissions, respectively.  
33

34 For some of the principal RFs (e.g., BC, land-use and mineral dust) there is not enough quantitative  
35 information available to assess their indirect effects, thus their RFs are the same as those presented in Table  
36 2.12. Table 2.5 gives the total (fossil and biomass burning) direct RFs for black carbon and organic carbon  
37 aerosols that are used to obtain the average shown in Figure 2.21. Table 2.13 summarizes the direct and  
38 indirect RFs presented in Figure 2.21, including the methods used for estimating the RFs and the associated  
39 uncertainty. Note that for indirect effects through changes in chemically active gases (e.g. OH or ozone) the  
40 emission based RF is not uniquely defined since the effect of one precursor will be affected by the levels of  
41 the other precursors. The RFs of indirect effects on methane and ozone by  $\text{NO}_x$ ,  $\text{CO}$  and VOC emissions are  
42 estimated by removing the anthropogenic emissions of one precursor at a time. Sensitivity analysis by  
43 Shindell et al. (2005) indicates that the non-linear effect induced by treating the precursors separately is of  
44 the order of 10% or less. Very uncertain indirect effects are not included in Table 2.13 and Figure 2.21.  
45 These include ozone changes due to solar effects, changes in secondary organic aerosols through changes in  
46 the  $\text{O}_3/\text{OH}$  ratio and apportioning of the cloud albedo changes to each aerosol type (Hansen et al., 2005).  
47

48 [INSERT FIGURE 2.21 HERE]

49  
50 [INSERT TABLE 2.13 HERE]

### 51 **2.9.4 Future Climate Impact of Current Emissions**

52  
53  
54 The changes in concentrations since pre-industrial time of the long-lived components causing the RF shown  
55 in Figure 2.20 are strongly influenced by the past-history of emissions. A different perspective is obtained by  
56 integrating RF over a future time horizon for a one year ‘pulse’ of global emissions (e.g., Jacobson, 2002  
57 used this approach to compare fossil fuel organic and black carbon aerosols to  $\text{CO}_2$ ). Comparing the

1 contribution from each forcing agent as shown in Figure 2.22 gives an indication of the future climate impact  
2 for current (year 2000) emissions of the different forcing agents. For the aerosols, the integrated RF is  
3 obtained based on the lifetimes, burdens, and RFs from the AEROCOM experiments, as summarized in  
4 Tables 2.4 and 2.5. For ozone precursors (CO, NO<sub>x</sub> and NMVOCs) data are taken from (Derwent et al.,  
5 2001; Collins et al., 2002; Stevenson et al., 2004; Berntsen et al., 2005), while for the long-lived species the  
6 radiative efficiencies and lifetimes are used, as well as a response function for CO<sub>2</sub> (see Table 2.14).  
7 Uncertainties in the estimates of the integrated RF originate both from uncertainties in lifetimes and optical  
8 properties as well as uncertainties in the current global emissions.  
9

10 [INSERT FIGURE 2.22 HERE]

11  
12 Figure 2.22 shows integrated RF for both a 20 and 100 year time horizon. Choosing the longer time horizon  
13 of 100 years, as was done in the GWPs for the long lived species included in the Kyoto Protocol, reduces the  
14 apparent importance of the shorter lived species. It should be noted that the compounds with long lifetimes  
15 and short emission histories will tend to contribute more to the total with this “forward looking” perspective  
16 than in the standard “IPCC RF bar-chart diagram” (Figure 2.20).  
17

### 18 2.9.5 Time Evolution of Radiative Forcing and Surface Forcing

19  
20 There is a good understanding of the time evolution of the LLGHG concentrations from in-situ  
21 measurements over the last few decades and extending further back using firn and ice-core data (see Section  
22 2.3, FAQ 2.1, Figure 1 and Chapter 6). Increases in RF are clearly dominated by CO<sub>2</sub>. Halocarbon RF has  
23 grown rapidly since 1950, but the RF growth has been cut dramatically by the Montreal Protocol (see  
24 Section 2.3.4). CFC RF is declining; in addition the combined RF of all ozone depleting substances (ODS)  
25 appears to have peaked at 0.32 W m<sup>-2</sup> during 2003. However, substitutes for ODS are growing at a slightly  
26 faster rate, so halocarbon RF growth is still positive (Table 2.1). Although the trend in halocarbon RF since  
27 the time of the TAR has been positive (see Table 2.1), the halocarbon RF in this report, as shown in Table  
28 2.12, is the same as in the TAR; this is due to a re-evaluation of the TAR results.  
29

30 RF timeseries for the natural (solar, volcanic aerosol) forcings are reasonably well known for the past 25  
31 years; estimates further back are prone to uncertainties (Section 2.7). Determining the timeseries for aerosol  
32 and ozone RF is far more difficult because of uncertainties in the knowledge of past emissions and chemical-  
33 microphysical modelling. Several timeseries for these and other RFs have been constructed (e.g.,  
34 Ramaswamy et al., 2001, Myhre et al., 2001; Hansen et al., 2002). GCMs develop their own time evolution  
35 of many forcings based on the temporal history of the relevant concentrations. As an example, the time  
36 evolution of the global-and-annual-mean, instantaneous, all-sky RF and *surface forcing* due to the principal  
37 agents in the MIROC+SPRINTARS GCM (Nozawa et al., 2005; Takemura et al., 2005) is illustrated in  
38 Figure 2.23. Although there are differences between models with regards to the temporal reconstructions and  
39 thus present-day forcing estimates, they typically have a qualitatively similar time-evolution since they often  
40 base the time histories on similar emissions data.  
41

42 GCMs compute the climate response based on the knowledge of the forcing agents and their time evolution.  
43 While most current GCMs incorporate the trace gas RFs, aerosol direct effects, solar and volcanoes, a few  
44 have in addition incorporated land-use change and cloud albedo effect. While LLGHGs have increased  
45 rapidly over the past 20 years and contribute the most to the present RF (refer also to Figure 2.20 and FAQ  
46 2.1, Figure 1), Figure 2.23 also indicates that the combined positive RF of the greenhouse gases exceeds the  
47 contributions due to all other anthropogenic agents throughout the latter half of the 20th century.  
48

49 The solar RF has a small positive value. The positive solar irradiance RF is *likely* to be at least five times  
50 smaller than the combined RF due to all anthropogenic agents, and about an order of magnitude less than the  
51 total greenhouse gas contribution (Figure 2.20, 2.23 and Table 2.12; see also the Foukal et al., 2006 review).  
52 The combined natural RF consists of the solar RF plus the large but transitory negative RF from episodic,  
53 explosive volcanic eruptions of which there have been several over the past half century (see Figure 2.18).  
54 Over particularly the 1950–2005 period, the combined natural forcing has been either negative or a small  
55 positive value (less than approximately 0.2 W m<sup>-2</sup>), reaffirming and extending the conclusions in the TAR.  
56 Therefore, it is *exceptionally unlikely* that natural RFs could have contributed a positive RF of comparable  
57 magnitude to the combined anthropogenic RF term over the period 1950-2005 (Figure 2.23). Attribution

1 studies with GCMs employ the available knowledge of the evolution of the forcing over the 20th century,  
2 and particularly the features distinguishing the anthropogenic from the natural agents (see also Chapter 9,  
3 Section 9.2).

4  
5 The *surface forcing* (Figure 2.23b), in contrast to RF, is dominated by the strongly negative shortwave effect  
6 of the aerosols (tropospheric and the episodic volcanic ones), with the LLGHGs exerting a small positive  
7 effect. Quantitative values of the RFs and *surface forcings* by the agents differ across models in view of the  
8 differences in model physics and in the formulation of the forcings due to the short-lived species (see  
9 Chapter 10 Section 10.2; also Collins et al. (2006) and Forster and Taylor (2006) for further discussion on  
10 uncertainties in GCMs calculation of RF and *surface forcing*). Again, as for RF, it is difficult to specify  
11 uncertainties in the time evolution, as emissions and concentrations for all but the LLGHGs are not well  
12 constrained.

13  
14 [INSERT FIGURE 2.23 HERE]

### 15 16 **2.9.6 Spatial Patterns of Radiative Forcing and Surface Forcing**

17  
18 Figure 6.7 of Ramaswamy et al. (2001) presented examples of the spatial patterns for most of the RF agents  
19 discussed in this chapter; these examples still hold. Many of the features seen in Figure 6.7 of Ramaswamy  
20 et al. (2001) are generic, although additional uncertainties exist, compared to uncertainties in the global-  
21 mean RF. Spatial patterns of the aerosol RF exhibit some of the largest differences between models,  
22 depending on the specification of the aerosols and their properties, and whether or not indirect cloud albedo  
23 effects are included. The aerosol direct and cloud-albedo effect RF also depend critically on the location of  
24 clouds, which differs between the GCMs. Illustrative examples of the spatial pattern of the instantaneous, RF  
25 between 1860 and present day, due to the natural plus anthropogenic agents, are presented from two GCMs  
26 in Figure 2.24. Volcanic aerosols play a negligible role in this calculation owing to the end-years considered  
27 and their virtual absence during these years. The MIROC+SPRINTARS model includes an aerosol cloud-  
28 albedo effect while the GFDL CM2.1 model (Delworth et al., 2005; Knutson et al., 2006) does not. RF over  
29 most of the globe is positive and is dominated by the LLGHGs. This is more so for the southern than the  
30 northern hemisphere, owing to the pronounced aerosol presence in the midlatitude northern hemisphere (see  
31 also Figure 2.12), with the regions of substantial aerosol RF clearly manifest over the source-rich continental  
32 areas. There are quantitative differences between the two GCMs in the global-mean RF, which are indicative  
33 of the uncertainties in the RF from the non-LLGHG agents, particularly aerosols (see Section 2.4 and Figure  
34 2.12d). The direct effect of aerosols is seen in the total RF of the GFDL model over northern hemisphere  
35 land regions, whereas the cloud albedo effect dominates the MIROC+SPRINTARS model in the  
36 stratocumulus low-latitude ocean regions. Note that the spatial pattern of the forcing is not indicative of the  
37 climate response pattern.

38  
39 [INSERT FIGURE 2.24 HERE]

40  
41 Wherever aerosol presence is considerable (namely the northern hemisphere), the *surface forcing* is negative,  
42 relative to preindustrial times (Figure 2.24). Because of the aerosol influence in the reduction of the  
43 shortwave radiation reaching the surface (see also Figure 2.12f), there is a net (sum of shortwave and  
44 longwave) negative *surface forcing* over a large part of the globe (see also Figure 2.23). In the absence of  
45 aerosols, LLGHGs increase the atmospheric longwave emission, with an accompanying increase in the  
46 longwave radiative flux reaching the surface. At high latitudes, and in parts of the southern hemisphere, there  
47 are fewer anthropogenic aerosols and thus, the *surface forcing* has a positive value, owing to the LLGHGs.

48  
49 These spatial patterns of RF and *surface forcing* imply different changes in the northern hemisphere equator-  
50 to-pole gradients for the surface and tropopause. This, in turn, implies different changes in the amount of  
51 energy absorbed by the troposphere in the low and high latitudes. The aerosol influences are also manifest in  
52 the difference between northern and southern hemisphere, for both RF and *surface forcing*.

## 53 54 **2.10 GWPs and Other Metrics for Comparing Different Emissions**

### 55 56 **2.10.1 Definition of an emission Metric and the GWP**

1 Multi-component abatement strategies to limit anthropogenic climate change need a framework and  
 2 numerical values for the trade-off between emissions of different forcing agents. The Global Warming  
 3 Potential (GWP) or other emission metrics provide a tool that can be used to operationalize comprehensive  
 4 and cost-effective policies (Article 3 of the United Nations Framework Convention on Climate Change  
 5 (UNFCCC)) in a decentralised manner so that multi-gas emitters (nations, industries) can compose  
 6 mitigation measures, according to a specified emission constraint, by allowing for substitution between  
 7 different climate agents. The metric formulation will differ depending on whether a long-term climate  
 8 change constraint has been set (e.g., Manne and Reichels, 2001) or no specific long-term constraint has been  
 9 agreed upon (as in the Kyoto Protocol). Either metric formulation requires knowledge of the contribution to  
 10 climate change from emissions of various components over time. The metrics assessed in this report are  
 11 purely physical based. However, it should be noted that many economists have argued that emission metrics  
 12 need also to account for economical dimensions of the problem they are intended to address (e.g., Manne and  
 13 Richels, 2001; Bradford, 2001; Godal, 2003; O'Neill, 2003). Substitution of gases within an international  
 14 climate policy with a long-term target that includes economic factors is discussed in Chapter 3 of IPCC  
 15 WGIII AR4. Metrics based on this approach will not be discussed in this report.

16  
 17 A very general formulation of an emission metric is given by (e.g., Kandlikar, 1996):

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

18  
 19 where  $I(\Delta C_i(t))$  is a function describing the impact (damage and benefit) of change in climate ( $\Delta C$ ) at time  $t$ .  
 20 The expression  $g(t)$  is a weighting function over time (e.g.,  $g(t) = e^{-kt}$  is a simple discounting giving short-  
 21 term impacts more weight) (Heal, 1997; Nordhaus, 1997;). The subscript  $r$  refers to a baseline emission path.  
 22 For two emission perturbations  $i$  and  $j$  the absolute metric values  $AM_i$  and  $AM_j$  can be calculated to provide a  
 23 quantitative comparison of the two emission scenarios. In the special case where the emission scenarios  
 24 consist of only one component (as for the assumed pulse emissions in the definition of GWP), the ratio  
 25 between  $AM_i$  and  $AM_j$  can be interpreted as a relative emission index for component  $i$  versus a reference  
 26 component  $j$  (as CO<sub>2</sub> in the case of GWP).

27  
 28 There are several problematic issues related to defining a metric based on the general formulation given  
 29 above (Fuglestedt et al., 2003). A major problem is to define appropriate impact functions, although there  
 30 have been some initial attempts to do this for a range of possible climate impacts (Hammit et al., 1996; Tol,  
 31 2002; den Elzen et al., 2005). Given that impact functions can be defined,  $AM$  calculations would require  
 32 regionally resolved climate change data (temperature, precipitation, winds, etc.) which would have to be  
 33 based on GCM results with their inherent uncertainties (Shine et al., 2005b). Other problematic issues  
 34 include the definition of the temporal weighting function  $g(t)$  and the baseline emission scenarios.

35  
 36 Due to these difficulties, the simpler and purely physical GWP index, based on the time integrated global  
 37 mean RF of a pulse emission of 1 kg of some compound ( $i$ ) relative to that of 1 kg of the reference gas CO<sub>2</sub>,  
 38 was developed (IPCC, 1990) and adopted for use in the Kyoto Protocol. The GWP of component  $i$  is defined  
 39 by

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

41 where TH is the time horizon,  $RF_i$  is the global mean RF of component  $i$ ,  $a_i$  is the RF per unit mass increase  
 42 in atmospheric abundance of component  $i$  (radiative efficiency),  $[C_i(t)]$  is the time-dependent abundance of  $i$ ,  
 43 and the corresponding quantities for the reference gas in the denominator. The numerator and denominator  
 44 are called the absolute global warming potential (AGWP) of  $i$  and  $r$  respectively. All GWPs given in this  
 45 report use CO<sub>2</sub> as the reference gas. The simplifications made to derive the standard GWP index include (i)  
 46 setting  $g(t) = 1$  (i.e., no discounting) up until the time-horizon (TH) and then  $g(t)=0$  thereafter, (ii) choosing a  
 47 1-kg pulse emission, (iii) defining the impact function,  $I(\Delta C)$ , to be the global mean RF, (iv) assuming that  
 48 the climate response is equal for all RF mechanisms, and (v) evaluating the impact relative to a baseline  
 49 equal to current concentrations (i.e., setting  $I(\Delta C_r(t)) = 0$ ). The criticisms of the GWP metric have focused  
 50 on all of these simplifications (e.g., O'Neill, 2000; Smith and Wigley, 2000; Bradford, 2001; Godal, 2003).

1 However, as long as there is no consensus on which impact function ( $I(\Delta C)$ ) and temporal weighting  
2 functions to use (both involve value judgements), it is difficult to assess the implications of the  
3 simplifications objectively (O'Neill, 2000; Fuglestedt et al., 2003).

4  
5 The adequacy of the GWP concept has been widely debated since its introduction (O'Neill, 2000;  
6 Fuglestedt et al., 2003). By its definition, two sets of emissions that are equal in terms of their total GWP  
7 weighted emissions will not be equivalent in terms of the temporal evolution of climate response  
8 (Fuglestedt et al., 2000; Smith and Wigley, 2000). Using a 100-year time horizon as in the Kyoto Protocol,  
9 the effect of current emissions reductions (e.g., during the first commitment period under the Kyoto  
10 Protocol) that contain a significant fraction of short-lived species (e.g., methane) will give less temperature  
11 reductions towards the end of the time horizon, compared to reductions in CO<sub>2</sub> emissions only. GWPs can  
12 really only be expected to produce identical changes in one measure of climate change – integrated  
13 temperature change following emissions impulses – and only under a particular set of assumptions (O'Neill,  
14 2000). The Global Temperature Potential (GTP) metric (see Section 2.10.4.2) provides an alternative  
15 approach by comparing global mean temperature change at the end of a given time horizon. Compared to the  
16 GWP, the GTP gives equivalent climate response at a chosen time, while putting much less emphasis on near  
17 term climate fluctuations caused by emissions of short-lived species (e.g., methane). However, as long as it  
18 has not been determined, neither scientifically, economically nor politically, what the proper time horizon for  
19 evaluating “dangerous anthropogenic interference in the climate system” should be, the lack of temporal  
20 equivalence does not invalidate the GWP concept or provide a guidance as how to replace it. Although it has  
21 several known shortcomings, a multi-gas strategy using GWPs is very likely to have advantages over a CO<sub>2</sub>-  
22 only strategy (O'Neill, 2003). Thus a GWP remains the recommended metric to compare future climate  
23 impacts of emissions of long-lived climate gases.

24  
25 Globally averaged GWPs have been calculated for short lived species, e.g., ozone precursors and absorbing  
26 aerosols (Fuglestedt et al., 1999; Derwent et al., 2001; Collins et al., 2002; Stevenson et al., 2004; Berntsen  
27 et al., 2005; Bond and Sun, 2005). There might be substantial co-benefits realized in mitigation actions  
28 involving short-lived species affecting climate and air pollutants (Hansen and Sato, 2004); however the  
29 effectiveness of the inclusion of short-lived forcing agents into international agreements is not clear (Rypdal  
30 et al., 2005). To assess the possible climate impacts of short-lived species and compare those with the  
31 impacts of the LLGHGs, a metric is needed. However, there are serious limitations with the use of global-  
32 mean GWPs for this purpose. While the GWPs of the LLGHGs do not depend on location and time of  
33 emissions, the GWPs for short-lived species will be regionally and time dependent. The different response in  
34 precipitation to an aerosol RF compared to a LLGHG RF also suggests that the GWP concept may be too  
35 simplistic when applied to aerosols.

### 36 37 **2.10.2 Direct GWPs**

38  
39 All GWPs depend on the AGWP for CO<sub>2</sub> (the denominator in the definition of the GWP). The AGWP of  
40 CO<sub>2</sub> again depends on the radiative efficiency for a small perturbation of CO<sub>2</sub> from the current level of about  
41 380 ppm. The radiative efficiency per kilogram CO<sub>2</sub> has been calculated using the same expression as for the  
42 CO<sub>2</sub> RF in Section 2.3.1, with an updated background CO<sub>2</sub> mixing ratio of 378 ppm. For a small perturbation  
43 from 378 ppm the RF is 0.01413 W m<sup>-2</sup> ppm<sup>-1</sup> (8.7% lower than the TAR value). The CO<sub>2</sub> response function  
44 (see Table 2.14) is based on an updated version of the Bern carbon-cycle model (Bern2.5CC), using a  
45 background CO<sub>2</sub> concentration of 378 ppm. The increased background concentration of CO<sub>2</sub> means that the  
46 airborne fraction of emitted CO<sub>2</sub> (Chapter 7, Section 7.3) is enhanced, contributing to an increase in the  
47 AGWP for CO<sub>2</sub>. The AGWP values for CO<sub>2</sub> for 20, 100, and 500 year time horizons are  $2.47 \times 10^{-14}$ ,  $8.69 \times$   
48  $10^{-14}$ , and  $28.6 \times 10^{-14}$  W m<sup>-2</sup> yr (kg(CO<sub>2</sub>))<sup>-1</sup>, respectively. The uncertainty in the AGWP for CO<sub>2</sub> is estimated  
49 to be ±15%, with equal contributions from the CO<sub>2</sub> response function and the RF calculation.

50  
51 Updated radiative efficiencies for well mixed-greenhouse gases are given in Table 2.14. Since the TAR  
52 radiative efficiencies have been reviewed by Montzka et al. (2003) and Velders et al. (2005). Gohar et al.  
53 (2004) and Forster et al. (2005) have investigated HFC compounds with up to 40% differences between  
54 earlier published results. Based on a variety of radiative transfer codes they found that uncertainties can be  
55 reduced to around 12% with well-constrained experiments. The HFCs studied were HFC-23, HFC-32, HFC-  
56 134a, and HFC-227ea. Hurley et al. (2005) studied the IR spectrum and RF of CF<sub>4</sub> and derived a 30% higher  
57 GWP value than given in the TAR. The RF calculations for GWPs for CH<sub>4</sub>, N<sub>2</sub>O, and halogen containing

1 well-mixed greenhouse gases employ the simplified formulas given in Ramaswamy et al. (2001) (see the  
2 TAR, Chapter 6, Table 6.2). Table 2.14 gives GWP values for time horizons of 20, 100 and 500 years. The  
3 species in Table 2.14 are those for which either significant concentrations or large trends in concentrations  
4 are observed, or a clear potential for future emissions has been identified. The uncertainties of these direct  
5 GWPs are taken to be  $\pm 35\%$  for the 5-95% (90%) confidence range.

6  
7 [INSERT TABLE 2.14 HERE]

### 8 9 **2.10.3 Indirect GWPs**

10 Indirect radiative effects include the direct effects of degradation products or the radiative effects of changes  
11 in concentrations of greenhouse gases caused by the presence of the emitted gas or its degradation products.  
12 Direct effects of degradation products for the greenhouse gases are not considered to be significant (WMO,  
13 2003). The indirect effects discussed here are linked to ozone formation or destruction, enhancement of  
14 stratospheric water vapour, changes concentrations of the OH radical with the main effect of changing the  
15 lifetime of methane, and secondary aerosol formation. Uncertainties for the indirect GWPs are generally  
16 much higher than for the direct GWPs. The indirect GWP will in many cases depend on the location and  
17 time of the emissions. For some species (e.g.,  $\text{NO}_x$ ) the indirect effects can be of opposite sign, further  
18 increasing the uncertainty of the net GWP. This can be because background levels of reactive species (e.g.,  
19  $\text{NO}_x$ ) can affect the chemical response non-linearly, and/or because the lifetime or the radiative effects of  
20 short lived secondary species formed can be regionally dependent. Thus the usefulness of the global mean  
21 GWPs to inform policy decisions can be limited. However, they are readily calculable and give an indication  
22 of the total potential of mitigating climate change by including a certain forcing agent in climate policy.  
23 Following the approach taken by the SAR and the TAR, the  $\text{CO}_2$  produced from oxidation of  $\text{CH}_4$ , CO and  
24 NMVOCs of fossil origin is not included in the GWP estimates since this carbon has been included in the  
25 national  $\text{CO}_2$  inventories. This issue may need to be reconsidered as inventory guidelines are revised.

#### 26 27 28 **2.10.3.1 Methane**

29 Four indirect radiative effects of methane emissions have been identified (see Prather et al., 2001;  
30 Ramaswamy et al., 2001). Methane enhances its own lifetime through changes in the OH concentration: it  
31 leads to changes in tropospheric ozone, it enhances stratospheric water vapour levels, and produces  $\text{CO}_2$ . The  
32 GWP given in Table 2.14 includes the first three of these effects. The lifetime effect is included by adopting  
33 a perturbation lifetime of 12 years (see Chapter 7, Section 7.4). The effect of ozone production is still  
34 uncertain, and as in the TAR it is included by enhancing the net of the direct and the lifetime effect by 25%.  
35 The estimate of RF caused by an increase in stratospheric water vapour has been increased significantly  
36 since the TAR (see Section 2.3.7). This has also been taken into account in the GWP estimate for methane by  
37 increasing the enhancement factor from 5% (TAR) to 15%. As a result the 100-yr GWP for methane has  
38 increased from 23 in the TAR to 25.

#### 39 40 41 **2.10.3.2 Carbon monoxide**

42 The indirect effects of carbon monoxide (CO) occur through reduced OH levels (leading to enhanced  
43 concentrations of methane) and enhancement of ozone. The TAR gave a range of 1.0–3.0 for the 100-yr  
44 GWP. Since the TAR, Collins et al. (2002) and Berntsen et al. (2005) have calculated GWPs for CO  
45 emissions, which range between 1.6 and 2.0, depending on the location of the emissions. Berntsen et al.  
46 (2005) found that emissions of CO from Asia had a 25% higher GWP compared to European emissions.  
47 Averaging over the TAR values and the new estimates give a mean of 1.9 for the 100-yr GWP for CO.

#### 48 49 50 **2.10.3.3 NMVOCs**

51 Collins et al. (2002) have calculated indirect GWPs for 10 non-methane volatile organic compounds  
52 (NMVOCs) with a global 3-D Lagrangian chemistry-transport model. Impacts on tropospheric ozone,  
53 methane (through changes in OH), and  $\text{CO}_2$  have been considered, using either an “anthropogenic” emission  
54 distribution or a “natural” emission distribution depending on the main sources for each gas. The indirect  
55 GWP values are given in Table 2.15. Weighting these GWPs by the emissions of the respective compounds  
56 gives a weighted average 100-yr GWP of 3.4. 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 of nitrate aerosol formation (see Section 2.4.4.5), which has not yet been included in model studies calculating GWPs for  $NO_x$ , can be significant. Due to the non-linear chemistry, the net RF of  $NO_x$  emissions will depend strongly on the location of emission and, with a strict definition of a pulse emission for the GWP, also on timing (daily, seasonal) of the emissions (Fuglestedt 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  $NO_x$  among the different studies and the omission of the nitrate aerosol effect, a central estimate for the 100-yr GWP for  $NO_x$  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 based on observed ozone depletion between 1980 and 1990 for 2005 emissions 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 an indirect 100-yr GWP for the tropospheric effects of  $H_2$  of 5.8, which includes the effects of methane lifetime and tropospheric ozone.

### 2.10.4 *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; Fuglestedt 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.

#### 2.10.4.1 *Revised GWP Formulations*

##### 2.10.4.1.2 *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). Fuglestedt et al. (2003) have proposed a revised GWP concept that includes the efficacy of a forcing agent. 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.

##### 2.10.4.2 *The Global-Temperature Potential*

1 Shine et al. (2005a) have proposed a Global Temperature Potential (GTP) as a new relative emission metric.  
2 The GTP is defined as the ratio between the global mean surface temperature change at a given future time  
3 horizon (TH) following an emission (pulse or sustained) of a compound  $x$  relative to a reference gas  $r$  (e.g.,  
4  $\text{CO}_2$ ).

$$5 \quad GTP_x^{TH} = \frac{\Delta T_x^H}{\Delta T_r^H}$$

6 where  $\Delta T_x^H$  denotes the global mean surface temperature change after  $H$  years following an emission of  
7 compound  $x$ . The GTPs do not require simulations with AOGCMs, but are given as transparent and simple  
8 formulas that employ a small number of input parameters required for calculation. Note that while the GWP  
9 is an integral quantity over the time horizon (i.e., the RF at the beginning and end of the time horizon counts  
10 exactly equal), the GTP uses the temperature change at time  $H$  (i.e., RF closer to time  $H$  contributes  
11 relatively more). The GTP metric requires knowledge of the same parameters as the GWP metric (radiative  
12 efficiency and lifetimes), but in addition the response times for the climate system must be known, in  
13 particular, if the lifetime of component  $x$  is very different from the lifetime of the reference gas. Differences  
14 in climate efficacies can easily be incorporated into the GTP metric. Due to the inclusion of the response  
15 times for the climate system, the GTP values for pulse emissions of gases with shorter lifetimes than the  
16 reference gas will be lower than the corresponding GWP values. As noted by Shine et al. (2005a) there is a  
17 near equivalence between the GTP for sustained emission changes and the pulse GWP. The GTP metric has  
18 the potential advantage over GWP that it is more directly related to surface temperature change.  
19

1 **References**

- 2
- 3 Abdul-Razzak, H., and S.J. Ghan, 2002: A parameterization of aerosol activation: 3. Sectional  
4 representation. *J. Geophys. Res.*, **107**(D3), 4026, doi: 10.1029/2001JD000483.
- 5 Abel, S.J., J.M. Haywood, E.J. Highwood, J. Li, and P.R. Buseck, 2003: Evolution of biomass burning  
6 aerosol properties from an agricultural fire in southern Africa. *Geophys. Res. Lett.*, **30**(15), 1783,  
7 doi:10.1029/2003GL017342.
- 8 Abel, S.J., E.J. Highwood, J.M. Haywood, and M.A. Stringer, 2005: The direct radiative effect of biomass  
9 burning aerosol over southern Africa. *Atmos. Chem. Phys. Disc.*, **5**, 1165-1211.
- 10 Ackerman, A.S., et al., 2000a: Reduction of tropical cloudiness by soot. *Science*, **288**, 1042-1047.
- 11 Ackerman, A.S., et al., 2000b: Effects of aerosols on cloud albedo: evaluation of Twomey's parameterization  
12 of cloud susceptibility using measurements of ship tracks. *J. Atmos. Sci.*, **57**, 2684-2695.
- 13 Ackerman, A.S., M.P. Kirkpatrick, D.E. Stevens and O.B. Toon, 2004: The impact of humidity above  
14 stratiform clouds on indirect aerosol climate forcing. *Nature*, **432**, 1014-1017.
- 15 Adams, P.J., J.H. Seinfeld, D. Koch, L. Mickley, and D. Jacob, 2001: General circulation model assessment  
16 of direct radiative forcing by the sulfate-nitrate-ammonium-water inorganic aerosol system. *J.*  
17 *Geophys. Res.*, **106**(D1), 1097-1112.
- 18 Adams, P.J. and J.H. Seinfeld, 2002: Predicting global aerosol size distributions in general circulation  
19 models. *J. Geophys. Res.*, **107**(D19), 4370, doi:10.1029/2001JD001010.
- 20 Albrecht, B., 1989: Aerosols, cloud microphysics and fractional cloudiness. *Science*, **245**, 1227-1230.
- 21 Allan, W., D. Lowe, A. Gomez, H. Struthers, and G. Brailsford, 2005: Interannual variation of <sup>13</sup>C in  
22 tropospheric methane: Implications for a possible atomic chlorine sink in the marine boundary layer.  
23 *J. Geophys. Res.*, **110**, D11306, doi:10.1029/2004JD005650.
- 24 Ammann, C.M., G.A. Meehl, W.M. Washington, and C.S. Zender, 2003: A monthly and latitudinally  
25 varying volcanic forcing dataset in simulations of 20th century climate. *Geophys. Res. Lett.*, **30**(12),  
26 1657, doi:10.1029/2003GL016875.
- 27 Anderson, T. L., et al., 2003: Climate forcing by aerosols: a hazy picture. *Science*, **300**, 1103-1104.
- 28 Anderson, T.L., et al., 2005a. Testing the MODIS satellite retrieval of aerosol fine-mode fraction. *J.*  
29 *Geophys. Res.*, **110**, D18204, doi:10.1029/2005JD005978.
- 30 Anderson, T.L., et al., 2005b: An "A-Train" strategy for quantifying direct climate forcing by anthropogenic  
31 aerosols. *Bull. Am. Meteorol. Soc.*, **86**, 1795-1809
- 32 Andreae, M.O., and P. Merlet, 2001: Emission of trace gases and aerosols from biomass burning. *Global*  
33 *Biogeochem. Cycles*, **15**(4), 955-966, 10.1029/2000GB001382.
- 34 Andreae, M.O., et al. 2004: Atmospheric science: smoking rain clouds over the Amazon. *Science*, **303**,  
35 5662,1337-1341.
- 36 Andres, R.J., G. Marland, T. Boden, and S. Bischof, 2000: Carbon dioxide emissions from fossil fuel  
37 consumption and cement manufacture, 1751-1991, and an estimate of their isotopic composition and  
38 latitudinal distribution. In: *The Carbon Cycle* [Wigley, T.M.L., and D.S. Schimel (eds.)]. Cambridge  
39 University Press, Cambridge, UK, pp. 53-62.
- 40 Andronova, N.G., E.V. Rozanov, F. Yang, M.E. Schlesinger, and G.L. Stenchikov, 1999: Radiative forcing  
41 by volcanic aerosols from 1850 to 1994. *J. Geophys. Res.*, **104**(D14), 16,807-16,826.
- 42 Antuña, J.C., et al., 2003: Spatial and temporal variability of the stratospheric aerosol cloud produced by the  
43 1991 Mount Pinatubo eruption. *J. Geophys. Res.*, **108**, 4624, doi:10.1029/2003JD003722.
- 44 Asano, S., A. Uchiyama, A. Yamazaki, J.-F. Gayet, and M. Tanizono, 2002: Two case studies of winter  
45 continental-type water and mixed-phased stratocumuli over the sea. II: Absorption of solar radiation.  
46 *J. Geophys. Res.*, **107**(D21), 4570, doi: 10.1029/?2001JD001108.
- 47 Baliunas, S., and R. Jastrow, 1990: Evidence for long-term brightness changes of solar-type stars. *Nature*,  
48 **348**, 520-522.
- 49 Balkanski, Y., M. Schulz, T. Claquin, C. Moulin, and P. Ginoux, 2004: Global emissions of mineral aerosol:  
50 formulation and validation using satellite imagery. In: *Emission of Atmospheric Trace Compounds*  
51 [Granier, C., P. Artaxo, and C.E. Reeves (eds.)]. Kluwer, Amsterdam, pp. 239-267.
- 52 Bange, H.W., S. Rapsomanikis, and M.O. Andreae, 1996: Nitrous oxide in coastal waters. *Global*  
53 *Biogeochem. Cycles*, **10**(1), 197-207.
- 54 Baran, A.J., and J.S. Foot, 1994: A new application of the operational sounder HIRS in determining a  
55 climatology of sulfuric acid aerosol from the Pinatubo eruption. *J. Geophys. Res.*, **99**(D12), 25,673-  
56 25,679

- 1 Barnes, J.E., and D.J. Hoffman (1997), Lidar measurements of stratospheric aerosol over Mauna Loa  
2 Observatory. *Geophys. Res. Lett.*, **24**(15), 1923-1926
- 3 Barnston, A.G., and P.T. Schickedanz, 1984: The effect of irrigation on warm season precipitation in the  
4 southern Great Plains. *J. Appl. Meteorol.*, **23**, 865-888.
- 5 Bartholome, E. and Belward, A.S., 2005: GLC2000: a new approach to global land cover mapping from  
6 Earth observation data. *Int. J. Remote Sens.*, **26**, 1959-1977.
- 7 Bates, T.S., et al., 2006: Aerosol direct radiative effects over the northwest Atlantic, northwest Pacific, and  
8 North Indian Oceans: estimates based on in situ chemical and optical measurements and chemical  
9 transport modelling, *Atmos. Chem. Phys. Discuss.*, **6**, 175-362.
- 10 Bauer, S.E., and D. Koch, 2005: Impact of heterogeneous sulfate formation at mineral dust surfaces on  
11 aerosol loads and radiative forcing in the Goddard Institute for Space Studies general circulation  
12 model. *J. Geophys. Res.*, **110**(D17202), doi:10.1029/2005JD005870.
- 13 Bauman, J.J., P.B. Russell, M.A. Geller, and P. Hamill, 2003: A stratospheric aerosol climatology from  
14 SAGE II and CLAES measurements: 2. Results and comparisons, 1984-1999. *J. Geophys. Res.*,  
15 **108**(D13), 4383, doi:10.1029/2002JD002993.
- 16 Beheng, K.D., 1994: A parameterization of warm cloud microphysical conversion process. *Atmos. Res.*, **33**,  
17 193-206.
- 18 Bekki, S., K. Law, and J. Pyle, 1994: Effect of ozone depletion on atmospheric CH<sub>4</sub> and CO concentrations.  
19 *Nature*, **371**, 595-597.
- 20 Bellouin, B., O. Boucher, J. Haywood, and M.S. Reddy, 2005: Global estimates of aerosol direct radiative  
21 forcing from satellite measurements. *Nature*, **439**, 1138-1141.
- 22 Bellouin, N., O. Boucher, D. Tanré, and O. Dubovik, 2003: Aerosol absorption over the clear-sky oceans  
23 deduced from POLDER-1 and AERONET observations. *Geophys. Res. Lett.*, **30**(14), 1748, doi:  
24 10.1029/2003GL017121.
- 25 Benestad, R. E., 2005: A review of the solar cycle length estimates. *Geophys. Res. Lett.*, **32**, L15714,  
26 doi:10.1029/2005GL023621.
- 27 Bergamaschi, P., et al., 2005: Inverse modelling of national and European CH<sub>4</sub> emissions using the  
28 atmospheric zoom model TM5. *Atmos. Chem. Phys.*, **5**, 2431-2460.
- 29 Bergstrom, R.W., P. Pilewskie, B. Schmid, and P.B. Russell, 2003: Estimates of the spectral aerosol single  
30 scattering albedo and aerosol radiative effects during SAFARI 2000. *J. Geophys. Res.*, **108**(D13),  
31 8474, doi:10.1029/2002JD002435
- 32 Berntsen, T.K., et al., 2005: Climate response to regional emissions of ozone precursors; sensitivities and  
33 warming potentials. *Tellus*, **4B**, 283-304.
- 34 Berntsen, T.K., J.S. Fuglestedt, G. Myhre, F. Stordal, and T.F. Berglen, 2006: Abatement of greenhouse  
35 gases: does location matter? *Clim. Change* **74**, 377-411
- 36 Berry, E.X., 1967: Cloud droplet growth by collection. *J. Atmos. Sci.*, **24**, 688-701.
- 37 Betts, R.A., 2000: Offset of the potential carbon sink from boreal forestation by decreases in surface albedo.  
38 *Nature*, **408**, 6809,187-189.
- 39 Betts, R.A., 2001: Biogeophysical impacts of land use on present-day climate: near-surface temperature  
40 change and radiative forcing. *Atmos. Sci. Lett.*, **2**, (1-4), doi:1006/asle.2001.0023.
- 41 Betts, R.A., and M.J. Best, 2004: *Relative impact of radiative forcing, landscape effects and local heat*  
42 *sources on simulated climate change in urban areas*. BETWIXT Technical Briefing Note No. 6, Met  
43 Office, Exeter, U.K., 15 p.
- 44 Betts, R.A., P.M. Cox, S.E. Lee, and F.I. Woodward, 1997: Contrasting physiological and structural  
45 vegetation feedbacks in climate change simulations. *Nature*, **387**, 796-799.
- 46 Betts, R.A., et al., 2004: The role of ecosystem-atmosphere interactions in simulated Amazonian  
47 precipitation decrease and forest dieback under global climate warming. *Theor. App. Climatol.*, **78**,  
48 157-175
- 49 Betts, R.A., P.D. Falloon, K.K. Goldewijk, and N. Ramankutty, 2006: Biogeophysical effects of land use on  
50 climate: model simulations of radiative forcing and large-scale temperature change. *Agricultural and*  
51 *Forest Meteorology* (in press).
- 52 Bigler, M., et al., 2002: Sulphate record from a northeast Greenland ice core over the last 1200 years based  
53 on continuous flow analysis. *Ann.Glaciol.*, **35**, 250-256.
- 54 Bingen, C., D. Fussen, and F. Vanhellefont, 2004: A global climatology of stratospheric aerosol size  
55 distribution parameters derived from SAGE II data over the period 1984-2000: 2. Reference data. *J.*  
56 *Geophys. Res.*, **109**, D06202, doi:10.1029/2003JD003511.

- 1 Blake, D., and F. Rowland, 1988: Continuing worldwide increase in tropospheric methane, 1978 to 1987.  
2 *Science*, **239**, 1129-1131.
- 3 Blake, N.J., et al., 2001: Large-scale latitudinal and vertical distributions of NMHCs and selected  
4 halocarbons in the troposphere over the Pacific Ocean during the March-April 1999 Pacific  
5 Exploratory Mission (PEM-Tropics B). *J. Geophys. Res.*, **106**, 32627-32644.
- 6 Bluth, G.J.S., S.D. Doiron, A.J. Krueger, L.S. Walter, and C. C. Schnetzler, 1992: Global tracking of the  
7 SO<sub>2</sub> clouds from the June 1991 Mount Pinatubo eruptions. *Geophys. Res. Lett.*, **19**(2), 151-154,  
8 10.1029/91GL02792.
- 9 Bluth, G.J.S., W.I. Rose, I.E. Sprod, and A.J. Krueger, 1997: Stratospheric loading of sulfur from explosive  
10 volcanic eruptions. *J. Geol.*, **105**, 671-683.
- 11 Boberg, F. and H. Lundstedt, 2002: Solar wind variations related to fluctuations of the North Atlantic  
12 Oscillation. *Geophys. Res. Lett.*, **29**(15), doi:10.1029/2002GL014903.
- 13 Boer, G.J., and B. Yu, 2003a: Climate sensitivity and climate state. *Clim. Dyn.*, **21**, 167-176.
- 14 Boer, G.J., and B. Yu, 2003b: Climate sensitivity and response. *Clim. Dyn.*, **20**, 415-429.
- 15 Bond, T.C., 2001: Spectral dependence of visible light absorption by carbonaceous particles emitted from  
16 coal combustion. *Geophys. Res. Lett.*, **28**(21), 4075-4078.
- 17 Bond, T.C., and H. Sun, 2005: Can reducing black carbon emissions counteract global warming? *Environ.*  
18 *Sci. Technol.*, **39**, 5921-5926, 2005.
- 19 Bond, T.C., et al., 1999: Light absorption by primary particle emissions from a lignite burning plant.  
20 *Environ. Sci. Technol.*, **33**, 3887-3891.
- 21 Bond, T.C., et al., 2004: A technology-based global inventory of black and organic carbon emissions from  
22 combustion. *J. Geophys. Res.*, **109**, D14203, doi:10.1029/2003JD003697.
- 23 Born, M., H. Dorr, and I. Levin, 1990: Methane consumption in aerated soils of the temperate zone. *Tellus* ,  
24 **42B**, 2-8.
- 25 Bortz S.E., and M.J. Prather, 2006: Ozone, water vapor, and temperature in the upper tropical troposphere:  
26 Variations over a decade of MOZAIC Measurements. *J. Geophys. Res.*, **111**, D05305,  
27 doi:10.1029/2005JD006512.
- 28 Boucher, O., 1999: Air traffic may increase cirrus cloudiness. *Nature*, **397**, 30-31.
- 29 Boucher, O. and U. Lohmann, 1995: The sulfate-CCN-cloud albedo effect: a sensitivity study using two  
30 general circulation models. *Tellus*, **47B**, 281-300.
- 31 Boucher, O., and D. Tanré, 2000: Estimation of the aerosol perturbation to the Earth's radiative budget over  
32 oceans using POLDER satellite aerosol retrievals. *Geophys. Res. Lett.*, **27**(8), 1103-1106.
- 33 Boucher, O., and J. Haywood, 2001: On summing the components of radiative forcing of climate change.  
34 *Clim. Dyn.*, **18**, 297-302.
- 35 Boucher, O., and M. Pham, 2002: History of sulfate aerosol radiative forcings. *Geophys. Res. Lett.*, **29**(9),  
36 22-25.
- 37 Boucher, O., H. Le Treut, and M.B. Baker, 1995: Precipitation and radiation modeling in a general  
38 circulation model: introduction of cloud microphysical processes. *J. Geophys. Res.*, **100**(D8),  
39 16,395- 16,414.
- 40 Boucher, O., G. Myhre, and A. Myhre, 2004: Direct human influence of irrigation on atmospheric water  
41 vapour and climate. *Clim. Dyn.*, **22**, 597-604.
- 42 Bounoua, L., R. Defries, G.J. Collatz, P. Sellers, and H. Khan, 2002: Effects of land cover conversion on  
43 surface climate. *Clim. Change*, **52**, 29-64.
- 44 Bousquet, P., D.A. Hauglustaine, P. Peylin, C. Carouge, and P. Ciais, 2005: Two decades of OH variability  
45 as inferred by an inversion of atmospheric transport and chemistry of methyl chloroform. *Atmos.*  
46 *Chem. Phys.*, **5**, 1679-1731.
- 47 Bower, K.N., et al., 1994: A parameterization of warm clouds for use in atmospheric general circulation  
48 models. *J. Atmos. Sci.*, **51**, 2722-2732.
- 49 *BP Statistical Review of World Energy, 2006*. London : BP p.l.c., v.  
50 <http://www.bp.com/productlanding.do?categoryId=91&contentId=7017990>
- 51 Bradford, D.F., 2001: Global change: time, money and tradeoffs. *Nature*, **410**, 649-650, doi:  
52 10.1038/35070707
- 53 Brasseur, G., and C. Granier, 1992: Mount Pinatubo aerosols, chlorofluorocarbons and ozone depletion.  
54 *Science*, **257**, 1239-1242.
- 55 Brenguier, J.L., et al., 2000a: Radiative properties of boundary layer clouds: droplet effective radius versus  
56 number concentration. *J. Atmos. Sci.*, **57**, 803-821.

- 1 Brenguier, J.L., et al., 2000b: An overview of the ACE-2 CLOUDYCOLUMN closure experiment. *Tellus*,  
2 **52B**, 815-827.
- 3 Brenguier, J.L., H. Pawlowska, and L. Schuller, 2003: Cloud microphysical and radiative properties for  
4 parameterization and satellite monitoring of the indirect effect of aerosol on climate. *J. Geophys.*  
5 *Res.*, 108(D15), 8632, doi:10.1029/2002JD002682.
- 6 Bréon, F.-M., D. Tanré, and S. Generoso, 2002: Aerosol effect on cloud droplet size monitored from  
7 satellite. *Science*, **295**, 834-838.
- 8 Bréon, F.-M. and M. Doutriaux-Boucher, 2005: A comparison of cloud droplet radii measured from space,  
9 *IEEE Trans. Geosci. Remote. Sens.*, **43**, 1796-1805, doi:10.1109/TGRS.2005.852838.
- 10 Broccoli, A.J., et al., 2003: Twentieth-century temperature and precipitation trends in ensemble climate  
11 simulations including natural and anthropogenic forcing. *J. Geophys. Res.*, **108**(D24), 4798,  
12 doi:10.1029/2003JD003812.
- 13 Brock, C.A., et al., 2004: Particle characteristics following cloud-modified transport from Asia to North  
14 America. *J. Geophys. Res.*, **109**, D23S26, 1-17.
- 15 Brovkin, V. M. et al., 2004. Role of land cover changes for atmospheric CO<sub>2</sub> increase and climate change  
16 during the last 150 years. *Global Change Biol.*, **10**, 1253-1266
- 17 Brovkin, V., M. et al., 2006: Biogeophysical effects of historical land cover changes simulated by six earth  
18 system models of intermediate complexity. *Clim. Dyn.* **26**(6), 587-600.
- 19 Cakmur, R.V., et al., 2006: Constraining the magnitude of the global dust cycle by minimizing the difference  
20 between a model and observations. *J. Geophys. Res.*, **111**, doi:10.1029/2005JD005791
- 21 Campbell, J.R., et al., 2003: Micropulse Lidar observations of tropospheric aerosols over northeastern South  
22 Africa during the ARREX and SAFARI-2000 dry season experiments. *J. Geophys. Res.*, **108**(D13),  
23 8497, doi:10.1029/2002JD002563.
- 24 Carslaw, K.S., R.G. Harrison, and J. Kirkby, 2002: Atmospheric science: Cosmic rays, clouds, and climate.  
25 *Science*, **298**, 1732-1737.
- 26 Chapman, G. A., Cookson, A. M. and J.J. Dobias, 1996: Variations in total solar irradiance during solar cycle  
27 22. *J. Geophys. Res.*, **101**, 13541--13548.
- 28 Chase, T.N., R.A. Pielke, T.G.F. Kittel, N. Nemani, and S.W. Running, 2000: Simulated impacts of  
29 historical land cover changes on global climate in northern winter. *Clim. Dyn.*, **16**, 93-105.
- 30 Chen, Y.-H., and R.G. Prinn, 2005: Atmospheric modelling of high- and low-frequency methane  
31 observations: Importance of interannually varying transport. *J. Geophys. Res.* **110**, D10303, doi:  
32 10.1029/2004JD005542.
- 33 Chen, Y. and J.E. Penner, 2005: Uncertainty analysis of the first indirect aerosol effect. *Atmos. Chem and*  
34 *Phys.*, **5**, 2935-2948.
- 35 Chen, Y.-H. and R.G. Prinn, 2006: Estimation of atmospheric methane emissions between 1996-2001 using  
36 a 3D global chemical transport model. *J. Geophys. Res.*, **111**, D10307, doi:10.1029/2005JD006058.
- 37 Chiapello, I., C. Moulin, and J.M. Prospero, 2005: Understanding the long-term variability of African dust  
38 transport across the Atlantic as recorded in both Barbados surface concentrations and large-scale  
39 Total Ozone Mapping Spectrometer (TOMS) optical thickness. *J. Geophys. Res.*, **110**, D18S10,  
40 doi:10.1029/2004JD005132.
- 41 Chipperfield, M.P., W.J. Randel, G.E. Bodeker, and P. Johnston, 2003: Global ozone: past and future. In:  
42 *Scientific Assessment of Ozone Depletion : 2002 : pursuant to Article 6 of the Montreal Protocol on*  
43 *Substances that Deplete the Ozone Layer* [Ennis, C.A. (ed.)]. WMO, Geneva, pp. 4.1-4.90.
- 44 Chou, M.-D., P.-K. Chan, and M. Wang, 2002: Aerosol radiative forcing derived from SeaWiFS-retrieved  
45 aerosol optical properties. *J. Atmos. Sci.*, **59**, 748-757.
- 46 Christiansen, B., 1999: Radiative forcing and climate sensitivity: the ozone experience. *Q. J. R. Meteorol.*  
47 *Soc.*, **125**, 3011-3035.
- 48 Christopher, S.A., and J. Zhang, 2004: Cloud-free shortwave aerosol radiative effect over oceans: strategies  
49 for identifying anthropogenic forcing from Terra satellite measurements. *Geophys. Res. Lett.*, **31**,  
50 L18101, doi:10.1029/2004GL020510.
- 51 Christopher, S.A., J. Zhang, Y.J. Kaufman, and L. Remer, 2006: Satellite-based assessment of the top of the  
52 atmosphere anthropogenic aerosol radiative forcing over cloud-free oceans, *Geophys. Res. Letts.*,  
53 **111**, L15816, doi:10.1029/2005GL025535
- 54 Chu, D.A., et al., 2002: Validation of MODIS aerosol optical depth retrieval over land. *Geophys. Res. Lett.*,  
55 **29**(12), doi:10.1029/2001GL013205.

- 1 Chu, D. A., et al., 2005: Evaluation of aerosol properties over ocean from Moderate Resolution Imaging  
2 Spectroradiometer (MODIS) during ACE-Asia. *J. Geophys. Res.*, **110**, D07308,  
3 doi:10.1029/2004JD005208.
- 4 Chuang, C.C., and J.E. Penner, 1995: Effects of anthropogenic sulfate on cloud drop nucleation and optical  
5 properties. *Tellus*, **47B**, 566-577.
- 6 Chuang, C.C., Penner, J.E., Taylor, K.E., Grossman, A.S., and J.J. Walton, 1997: An assessment of the  
7 radiative effects of anthropogenic sulfate. *J. Geophys. Res.*, **102**(D3), 3761-3778.
- 8 Chuang, C.C., J.E. Penner, J.M. Prospero, K.E. Grant, G.H. Rau, and K. Kawamoto, 2002: Cloud  
9 susceptibility and the first aerosol indirect forcing: Sensitivity to black carbon and aerosol  
10 concentrations. *J. Geophys. Res.*, **107**(D21), 4564, doi:10.1029/2000JD000215.
- 11 Chung, C., V. Ramanathan, D. Kim, I.A. Podgorny, 2005: Global anthropogenic aerosol direct forcing  
12 derived from satellite and ground-based observations. *J. Geophys. Res.*, **110**, D24207,  
13 doi:10.1029/2005JD006356.
- 14 Chuang, P.Y., 2003: Measurement of the timescale of hygroscopic growth for atmospheric aerosols. *J.*  
15 *Geophys. Res.*, **108**, (D9), 4282, 5-13.
- 16 Chung, S.H., and J.H. Seinfeld, 2002: Global distribution and climate forcing of carbonaceous aerosols. *J.*  
17 *Geophys. Res.*, **107**, doi:10.1029/2001JD001397.
- 18 Chylek, P., and J. Wong, 1995: Effect of absorbing aerosols on global radiation budget. *Geophys. Res. Lett.*,  
19 **22**(8), 929-931.
- 20 Chylek, P., B. Henderson, M. Mishchenko, 2003: Aerosol radiative forcing and the accuracy of satellite  
21 aerosol optical depth retrieval. *J. Geophys. Res.*, **108**(D24), 4764, doi:10.1029/2003JD004044.
- 22 Clarke, A.D., et al., 2004: Size distributions and mixtures of dust and black carbon aerosol in Asian outflow:  
23 Physiochemistry and optical properties. *J. Geophys. Res.*, **109**, D15S09, doi:10.1029/2003JD004378.
- 24 Claquin, T., M. Schulz, and Y. Balkanski, 1999: Modeling the mineralogy of atmospheric dust, *J. Geophys.*  
25 *Res.*, **104**(D18), 22243-22256.
- 26 Coen, M.C., et al., 2004: Saharan dust events at the Jungfrauoch: detection by wavelength dependence of  
27 the single scattering albedo and first climatology analysis. *Atmos. Chem. and Phys.*, **4**, 2465-2480.
- 28 Collins, W.D., et al., 2001: Simulating aerosols using a chemical transport model with assimilation of  
29 satellite aerosol retrievals: Methodology for INDOEX. *J. Geophys. Res.*, **106**(D7), 7313-7336.
- 30 Collins, W.D., et al., 2006: Radiative forcing by well-mixed greenhouse gases: Estimates from climate  
31 models in the IPCC AR4. *J. Geophys. Res.*, **111**, D14317, doi:10.1029/2005JD006713.
- 32 Collins, W.J., R.G. Derwent, C.E. Johnson, and D.S. Stevenson, 2002: The oxidation of organic compounds  
33 in the troposphere and their global warming potentials. *Clim. Change*, **52**, 453-479.
- 34 Considine, D.B., J.E. Rosenfield, and E.L. Fleming, 2001: An interactive model study of the influence of the  
35 Mount Pinatubo aerosol on stratospheric methane and water trends. *J. Geophys. Res.*, **106**(D21),  
36 27711-27727.
- 37 Conway, T.J., et al., 1994: Evidence for interannual variability of the carbon cycle from the NOAA/CMDL  
38 sampling network. *J. Geophys. Res.*, **99**(D11), 22,831-22,855.
- 39 Cook, J., and E.J. Highwood, 2004: Climate response to tropospheric absorbing aerosols in an intermediate  
40 general-circulation model. *Q.J.R. Meteorol. Soc.*, **130**, 175-191.
- 41 Cooke, W.F., C. Liousse, H. Cachier, and J. Feichter, 1999: Construction of a 10 x 10 fossil fuel emission  
42 data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model.  
43 *J. Geophys. Res.*, **104**(D18), 22,137-22,162.
- 44 Cooke, W.F., V. Ramaswamy, and P. Kasibhatla, 2002: A general circulation model study of the global  
45 carbonaceous aerosol distribution. *J. Geophys. Res.*, **107**(D16), 10.1029/2001JD001274.
- 46 Coughlin, K., and K.K. Tung, 2004: Eleven-year solar cycle signal throughout the lower atmosphere. *J.*  
47 *Geophys. Res.*, **109**, D21105, doi:10.1029/2004JD004873.
- 48 Cox, P.M., et al., 1999: The impact of new land surface physics on the GCM simulation of climate and  
49 climate sensitivity. *Clim. Dyn.* **15**, 183-203
- 50 Cramer, W., et al., 2001: Global response of terrestrial ecosystem structure and function to CO<sub>2</sub> and climate  
51 change: Results from six dynamic global vegetation models. *Global Change Biol* **7**, 357-373.
- 52 Crooks, S.A., and L.J. Gray, 2005: Characterization of the 11-year solar signal using a multiple regression  
53 analysis of the ERA-40 dataset. *J. Climate*, **18**, 996-1015.
- 54 Crutzen, P.J., 2004: New directions: the growing urban heat and pollution "island" effect - impact on  
55 chemistry and climate. *Atmos. Env.*, **38**, 3539-3540.

- 1 Cunnold, D.M., et al., 2002: In situ measurements of atmospheric methane at GAGE/AGAGE sites during  
2 1985-2000 and resulting source inferences. *J. Geophys. Res.*, **107**(D14), doi:  
3 10.1029/2001JD001226.
- 4 Cunnold, D.M. et al., 2004: Comment on “Enhanced upper stratospheric ozone: Sign of recovery or solar cycle  
5 effect?” by W. Steinbrecht et al. *J. Geophys. Res.*, **109**, D14305, doi:10.1029/2004JD004826.
- 6 Cziczo, D., D. Murphy, H.P., and D. Thompson, 2004: Single particle measurements of the chemical  
7 composition of cirrus ice residue from CRYSTAL-FACE. *J. Geophys. Res.*, **109**, D04201,  
8 doi:10.1029/2004JD004032.
- 9 Dameris, M., et al., 2005: Long-term changes and variability in a transient simulation with a chemistry-  
10 climate model employing realistic forcing. *Atmos. Chem. Phys. Disc.*, **5**, 2121-2145.
- 11 Daniel, J. S., S. Solomon, and D.L. Albritton, 1995: On the evaluation of halocarbon radiative forcing and  
12 global warming potentials. *J. Geophys. Res.*, **100**(D1), 1271-1286, 10.1029/94JD02516
- 13 de Ridder, K., and H. Gallée, 1998: Land surface-induced regional climate change in southern Israel. *J. Appl.*  
14 *Meteorol.*, **37**, 1470-1485.
- 15 Decesari, S., M.C. Facchini, S. Fuzzi, and E. Tagliavini, 2000: Characterization of water-soluble organic  
16 compounds in atmospheric aerosol: a new approach. *J. Geophys. Res.*, **105**(D1), 1481-1489.
- 17 Decesari, S., et al., 2001: Chemical features and seasonal variation of fine aerosol water-soluble organic  
18 compounds in the Po Valley, Italy. *Atmos. Env.*, **35**, 3691-3699.
- 19 Del Genio, A.D., M.S. Yao, and K.-W. Lo, 1996: A prognostic cloud water parameterization for global  
20 climate models. *J. Climate*, **9**, 270-304.
- 21 Delworth, T.L., V. Ramaswamy, and G.L. Stenchikov, 2005: The impact of aerosols on simulated ocean  
22 temperature and heat content in the 20th century. *Geophys. Res. Lett.* **32**, L24709,  
23 doi:10.1029/2005GL024457,
- 24 Dentener, F., et al., 2006: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 -  
25 prescribed data-sets for AeroCom. *Atmos. Chem. Phys. Disc.* **6**, 2703-2763.
- 26 Derwent, R.G., W.J. Collins, C.E. Johnson, and D.S. Stevenson, 2001: Transient behaviour of tropospheric  
27 ozone precursors in a global 3-D CTM and their indirect greenhouse effects. *Clim. Change*, **49**, 463-  
28 487.
- 29 Deshler, T., et al., 2003: Thirty years of in situ stratospheric aerosol size distribution measurements from  
30 Laramie, Wyoming (41N), using balloonborne instruments. *J. Geophys. Res.*, **108**(D5), 4167,  
31 doi:10.1029/2002JD002514.
- 32 Deuzé, J.L., et al., 2000: Estimate of the aerosol properties over the ocean with POLDER. *J. Geophys. Res.*,  
33 **105**, 15329-15346.
- 34 Deuzé, J.L., et al., 2001: Remote sensing of aerosols over land surfaces from POLDER-ADEOS-1 polarized  
35 measurements. *J. Geophys. Res.*, **106**, 4913-4926.
- 36 Dewitte, S., D. Crommelynck, S. Mekaoui, and A. Joukoff, 2005: Measurement and uncertainty of the long-  
37 term total solar irradiance trend. *Sol. Phys.*, **224**, 209-216.
- 38 Diab, R. D., A.M. Thompson, K. Mari, L. Ramsay, and G.J.R. Coetzee, 2004: Tropospheric ozone  
39 climatology over Irene, South Africa, from 1990 to 1994 and 1998 to 2002. *J. Geophys. Res.*, **109**,  
40 D20301, doi:10.1029/2004JD004793.
- 41 Dlugokencky, E.J., et al., 1996: Changes in CH<sub>4</sub> and CO growth rates after the eruption of Mt. Pinatubo and  
42 their link with changes in tropical tropospheric UV flux. *Geophys. Res. Lett.*, **23**(20), 2761-2764.
- 43 Dlugokencky, E.J., K.A. Masarie, P.M. Lang, and P.P. Tans, 1998: Continuing decline in the growth rate of  
44 atmospheric methane. *Nature*, **393**, 447-450.
- 45 Dlugokencky, E.J., B.P. Walter, K.A. Masarie, P.M. Lang, and E.S. Kasischke, 2001: Measurements of an  
46 anomalous global methane increase during 1998. *Geophys. Res. Lett.*, **28**(3), 499-502.
- 47 Dlugokencky, E.J., et al., 2003: Atmospheric methane levels off: Temporary pause or a new steady-state?  
48 *Geophys. Res. Lett.*, **30**(19), DOI: 10.1029/2003GL018126.
- 49 Dlugokencky, E.J., et al., 2005: Conversion of NOAA CMDL atmospheric dry air CH<sub>4</sub> mole fractions to a  
50 gravimetrically prepared standard scale. *J. Geophys. Res.*, **110**, D18306, doi:10.1029/2005JD006035.
- 51 Döll, P., 2002: Impact of climate change and variability on irrigation requirements: a global perspective.  
52 *Clim. Change*, **54**, 269-293.
- 53 Douglass, A., M. Schoeberl, R. Rood, and S. Pawson, 2003: Evaluation of transport in the lower tropical  
54 stratosphere in a global chemistry and transport model. *J. Geophys. Res.*, **108**, D9, 4259,  
55 doi:10.1029/2002JD002696
- 56 Douglass, D.H., and B.D. Clader, 2002: Climate sensitivity of the Earth to solar irradiance. *Geophys. Res.*  
57 *Lett.*, **29**(16), 33-36.

- 1 Dubovik, O., and M.D. King, 2000: A flexible inversion algorithm for retrieval of aerosol optical properties  
2 from Sun and sky radiance measurements. *J. Geophys. Res.*, **105**, 20673-20696.
- 3 Dubovik, O., et al., 1998: Single-scattering albedo of smoke retrieved from the sky radiance and solar  
4 transmittance measured from ground. *J. Geophys. Res.*, **103**(D24), 31,903-31,923.
- 5 Dubovik, O., et al., 2000: Accuracy assessments of aerosol optical properties retrieved from Aerosol Robotic  
6 Network (AERONET) Sun and sky radiance measurements. *J. Geophys. Res.*, **105**, 9791-9806.
- 7 Dubovik, O., et al., 2002: Variability of absorption and optical properties of key aerosol types observed in  
8 worldwide locations. *J. Atmos. Sci.*, **59**, 590-608.
- 9 Dufresne, L.-L., J. Quaas, O. Boucher, S. Denzil and L. Fairhead, 2005: Contrasts in the effects on climate  
10 of anthropogenic sulfate aerosols between the 20<sup>th</sup> and the 21<sup>st</sup> century. *Geophys. Res. Lett.*, **32**,  
11 L21703, doi: 10.1029/2005GL023619.
- 12 Duncan, B.N., et al., 2003: Indonesian wildfires of 1997: Impact on tropospheric chemistry. *J. Geophys.*  
13 *Res.*, **108**(D15), 4458, , doi:10.1029/2002JD003195.
- 14 Dusek, U., et al., 2006: Size matters more than chemistry for cloud-nucleating ability of aerosol particles.  
15 *Science*, **312**, 1375-1378.
- 16 Dziembowski, W.A., P.R. Goode, and J. Schou, 2001: Does the sun shrink with increasing magnetic  
17 activity? *Astrophys. J.*, **553**, 897-904.
- 18 Eagan, R., P.V. Hobbs, and L. Radke, 1974: Measurements of CCN and cloud droplet size distributions in the  
19 vicinity of forest fires. *J. Appl. Meteorol.*, **13**, 537-553.
- 20 Easter, R. et al., 2004: MIRAGE: Model description and evaluation of aerosols and trace gases. *J. Geophys.*  
21 *Res.* **109**, D20210, doi:10.1029/2004JD004571.
- 22 Eatough, D.J., et al., 2003: Semivolatile particulate organic material in southern Africa during SAFARI  
23 2000. *J. Geophys. Res.*, **108**(D13), 8479, doi:10.1029/2002JD002296.
- 24 Eck, T.F., et al., 2003: Variability of biomass burning aerosol optical characteristics in southern Africa  
25 during the SAFARI 2000 dry season campaign and a comparison of single scattering albedo  
26 estimates from radiometric measurements. *J. Geophys. Res.*, **108**(D13), 8477,  
27 doi:10.1029/2002JD002321.
- 28 Edwards, D.P., et al., 2004: Observations of carbon monoxide and aerosols from the Terra satellite: Northern  
29 Hemisphere variability. *J. Geophys. Res.*, **109**, D24202, doi:10.1029/2004JD004727.
- 30 Ehhalt, D.H., 1999: Gas phase chemistry of the troposphere. In: *Global Aspects of Atmospheric Chemistry*,  
31 *Vol. 6* [Baumgaertel, H., W. Gruenbein, and F. Hensel (eds.)]. Springer Verlag, Darmstadt, pp. 21-  
32 110.
- 33 den Elzen, M., et al., 2005. Analysing countries' contribution to climate change: Scientific and policy-related  
34 choices. *Environ. Sci. Technol.*, **8** (6), 614-636.
- 35 Erlick, C., L.M. Russell, and V. Ramaswamy, 2001: A microphysics-based investigation of the radiative  
36 effects of aerosol-cloud interactions for two MAST Experiment case studies. *J. Geophys. Res.*,  
37 **106**(D1), 1249-1269.
- 38 Ervens, B., G. Feingold and S.M. Kreidenweis, 2005: The influence of water-soluble organic carbon on  
39 cloud drop number concentration. *J. Geophys. Res.*, **110**, D18211, doi:10.1029/2004JD005634.
- 40 Etheridge, D.M., et al., 1996: Natural and anthropogenic changes in atmospheric CO<sub>2</sub> over the last 1000  
41 years from air in Antarctic ice and firn. *J. Geophys. Res.*, **101**(D2), 4115-4128.
- 42 Etheridge, D.M., L.P. Steele, R.J. Francey, and R.L. Langenfelds, 1998: Atmospheric methane between 1000  
43 A.D. and present: Evidence of anthropogenic emissions and climatic variability. *J. Geophys. Res.*,  
44 **103**(D13), 15,979-15,993.
- 45 Facchini, M.C., M. Mircea, S. Fuzzi, and R.J. Charlson, 1999: Cloud albedo enhancement by surface-active  
46 organic solutes in growing droplets. *Nature*, **401**, 257-259.
- 47 Facchini, M.C., S. Decesari, M. Mircea, S. Fuzzi, and G. Loglio, 2000: Surface tension of atmospheric wet  
48 aerosol and cloud/fog droplets in relation to their organic carbon content and chemical composition.  
49 *Atmos. Env.*, **34**, 4853-4857.
- 50 Feichter, J., E. Roeckner, U. Lohmann, and B. Liepert, 2004: Nonlinear aspects of the climate response to  
51 greenhouse gas and aerosol forcing. *J. Climate*, **17**, 2384-2398.
- 52 Feingold, G., 2003: Modelling of the first indirect effect: Analysis of measurement requirements. *Geophys.*  
53 *Res. Lett.*, **30**(19), 1997, doi:10.1029/2003GL017967.
- 54 Feingold, G., and P.Y. Chuang, 2002: Analysis of influence of film-forming compounds on droplet growth:  
55 Implications for cloud microphysical processes and climate. *J. Atmos. Sci.*, **59**, 2006-2018.
- 56 Feingold, G., R. Boers, B. Stevens and W.R. Cotton, 1997: A modelling study of the effect of drizzle on  
57 cloud optical depth and susceptibility. *J. Geophys. Res.*, **102**(D12), 13527-13534.

- 1 Feingold, G., W.L. Eberhard, D.E. Veron, and M. Previdi, 2003: First measurements of the Twomey indirect  
2 effect using ground-based remote sensors. *Geophys. Res. Lett.*, **30**(6), 1287,  
3 doi:10.1029/2002GL016633.
- 4 Feng, Y., and J. Penner. 2006: Global modeling of nitrate and ammonium: Interaction of aerosols and  
5 tropospheric chemistry. *J. Geophys. Res.* (in press)
- 6 Ferretti, D.F., D.C. Lowe, R.J. Martin, and G.W. Brailsford, 2000: A new GC-IRMS technique for high  
7 precision, N<sub>2</sub>O-free analysis of  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  in atmospheric CO<sub>2</sub> from small air samples. *J.*  
8 *Geophys. Res.*, **105**(D5), 6709-6718.
- 9 Field, C.B., R.B. Jackson, and H.A. Mooney, 1995: Stomatal responses to increased CO<sub>2</sub>: implications from  
10 the plant to the global scale. *Plant Cell Environ.*, **18**, 1214-1225.
- 11 Fioletov, V.E., G.E. Bodeker, A.J. Miller, R.D. McPeters, and R. Stolarski, 2002: Global and zonal total  
12 ozone variations estimated from ground-based and satellite measurements: 1964-2000. *J. Geophys.*  
13 *Res.*, **107**(D22), 4647, doi:10.1029/2001JD001350.
- 14 Formenti, P., et al., 2003: Inorganic and carbonaceous aerosols during the Southern African Regional  
15 Science Initiative (SAFARI 2000) experiment: Chemical characteristics, physical properties, and  
16 emission data or smoke from African biomass burning. *J. Geophys. Res.*, **108**(D13), 8488,  
17 doi:10.1029/2002JD002408, 2003.
- 18 Forster, P.M.F., and K.P. Shine, 1999: Stratospheric water vapour changes as a possible contributor to  
19 observed stratospheric cooling. *Geophys. Res. Lett.*, **26**(21), 3309-3312.
- 20 Forster, P.M.F., and K. Tourpali, 2001: Effect of tropopause height changes on the calculation of ozone  
21 trends and their radiative forcing. *J. Geophys. Res.*, **106**(D11), 12241-12251.
- 22 Forster, P.M.F., and K.E. Taylor, 2006: Climate forcings and climate sensitivities diagnosed from coupled  
23 climate model integrations. *J. Climate* (in press).
- 24 Forster, P.M.F., M. Blackburn, R. Glover, and K.P. Shine, 2000: An examination of climate sensitivity for  
25 idealised climate change experiments in an intermediate general circulation model. *Clim. Dyn.*, **16**,  
26 833-849.
- 27 Forster, P.M.F., and K.P. Shine, 2002: Assessing the climate impact of trends in stratospheric water vapor.  
28 *Geophys. Res. Lett.*, **29**(6), doi:10.1029/2001GL013909.
- 29 Forster, P.M.F., and M.J. Joshi, 2005: The role of halocarbons in the climate change of the troposphere and  
30 stratosphere. *Clim. Change*, **70**, 249-266.
- 31 Forster, P.M.F., et al., 2005: Resolving uncertainties in the radiative forcing of HFC-134s. *J. Quant.*  
32 *Spectrosc. Ra.*, **93**, 447-460.
- 33 Foster, S.S., 2004: *Reconstruction of solar irradiance variations for use in studies of global climate change:*  
34 *application of recent SOHO observations with historic data from the Greenwich Observatory.* PhD  
35 Thesis, University of Southampton, Faculty of Science, Southampton, 231 p.
- 36 Foukal, P., C. Frohlich, H. Spruit, and T. M. L. Wigley, 2006: Variations in solar luminosity and their effect  
37 on the Earth's climate. *Nature*, **443**, 161-166.
- 38 Francey, R.J., et al., 1995: Changes in oceanic and terrestrial carbon uptake since 1982. *Nature*, **373**, 326-  
39 330.
- 40 Francey, R.J., et al., 1999: A history of  $\delta^{13}\text{C}$  in atmospheric CH<sub>4</sub> from the Cape Grim Air Archive and  
41 Antarctic firn air. *J. Geophys. Res.*, **104**(D19), 23,633-23,643.
- 42 Frankenberg, C., J.F. Meirink, M. van Weele, U. Platt and T. Wagner, 2005: Assessing methane emissions  
43 from global space-borne observations. *Science*, **308**, 1010-1014.
- 44 Free, M., and J. Angell, 2002: Effect of volcanoes on the vertical temperature profile in radiosonde data. *J.*  
45 *Geophys. Res.*, **107**(D10), doi:10.1029/2001JD001128.
- 46 Friedl, M.A., et al., 2002: Global land cover mapping from MODIS: algorithms and early results. *Remote*  
47 *Sens. Env.*, **83**, 287-302.
- 48 Fröhlich, C., and J. Lean, 2004: Solar radiative output and its variability: Evidence and mechanisms. *Astron.*  
49 *Astrophys. Rev.*, **12**, 273-320.
- 50 Fuglestedt, J.S., et al., 1999: Climatic forcing of nitrogen oxides through changes in tropospheric ozone and  
51 methane; global 3D model studies. *Atmos. Env.*, **33**, 961-977.
- 52 Fuglestedt, J. S., T. K. Berntsen, O. Godal, and T. Skodvin, 2000: Climate implications of GWP-based  
53 reductions in greenhouse gas emissions. *Geophys. Res. Lett.*, **27**(3), 409-412,  
54 10.1029/1999GL010939.
- 55 Fuglestedt, J.S. et al., 2003: Metrics of climate change: assessing radiative forcing and emission indices.  
56 *Clim. Change*, **58**, 267-331.

- 1 Fueglistaler, S., H. Wernli, and T. Peter, 2004: Tropical troposphere-to-stratosphere transport inferred from  
2 trajectory calculations. *J. Geophys. Res.*, **109**, D03108.
- 3 Fuller, K.A., W.C. Malm, and S.M. Kreidenweis, 1999: Effects of mixing on extinction by carbonaceous  
4 particles. *J. Geophys. Res.*, **104**(D13), 15,941-15,954.
- 5 Fuzzi S, et al., 2001; A simplified model of the water soluble organic component of atmospheric aerosols.  
6 *Geophys. Res. Lett.*, **28**(21), 4079-4082
- 7 Gao, F., et al., 2005: MODIS bidirectional reflectance distribution function and albedo Climate Modeling  
8 Grid products and the variability of albedo for major global vegetation types. *J. Geophys. Res.*, **110**,  
9 D01104, doi:10.1029/2004JD005190.
- 10 Gauss, M., et al., 2006: Radiative forcing since preindustrial times due to ozone changes in the troposphere  
11 and the lower stratosphere. *Atmos. Chem. Phys.*, **6**, 575-599.
- 12 Gedney, N., et al., 2006: Detection of a direct carbon dioxide effect in continental river runoff records.  
13 *Nature*, **439**, 835-838, doi:10.1038/nature04504
- 14 Geller, M.A., and S.P. Smyshlyaev, 2002: A model study of total ozone evolution 1979-2000: The role of  
15 individual natural and anthropogenic effects. *Geophys. Res. Lett.*, **29**(22), 5-8.
- 16 Geogdzhayev, I.V., M.I. Mishchenko, W.B. Rossow, B. Cairns, and A.A. Lacis, 2002: Global two-channel  
17 AVHRR retrievals of aerosol properties over the ocean for the period of NOAA-9 observations and  
18 preliminary retrievals using NOAA-7 and NOAA-11 data. *J. Atmos. Sci.*, **59**, 262-278.
- 19 Ghan, S., et al., 2001: Evaluation of aerosol direct radiative forcing in MIRAGE. *J. Geophys. Res.*, **106**(D6),  
20 5295-5316.
- 21 Gillett, N.P., M.F. Wehner, S.F.B. Tett, and A.J. Weaver, 2004: Testing the linearity of the response to  
22 combined greenhouse gas and sulfate aerosol forcing. *Geophys. Res. Lett.*, **31**, L14201,  
23 doi:10.1029/2004GL020111.
- 24 Gleisner, H., and P. Thejll, 2003: Patterns of tropospheric response to solar variability. *Geophys. Res. Lett.*,  
25 **30**(13), 1711, doi:10.1029/2003GL017129, 2003.
- 26 Godal O., 2003: The IPCC assessment of multidisciplinary issues: The choice of greenhouse gas indices.  
27 *Clim. Change*, **58**, 243-249.
- 28 Gohar, L.K., G. Myhre, and K.P. Shine, 2004: Updated radiative forcing estimates of four halocarbons. *J.*  
29 *Geophys. Res.*, **109**, D01107, doi:10.1029/2003JD004320.
- 30 Goloub, P., and O. Arino, 2000: Verification of the consistency of POLDER aerosol index over land with  
31 ATSR-2/ERS-2 fire product. *Geophys. Res. Lett.*, **27**(6), 899-902.
- 32 Goloub, P., et al., 1999: Validation of the first algorithm applied for deriving the aerosol properties over the  
33 ocean using the POLDER/ADEOS measurements. *IEEE Trans. Remote Sens.*, **37**, 1586-1596.
- 34 Goosse, H., H. Renssen, A. Timmermann, and R.S. Bradley, 2005: Internal and forced climate variability  
35 during the last millennium: a model-data comparison using ensemble simulations. *Quaternary Sci.*  
36 *Rev.* **24**, 134-1360
- 37 Gordon, L.J., et al., 2005: Human modification of global water vapor flows from the land surface. *Proc.*  
38 *Natl. Acad. Sci. U.S.A.*, **102**, 7612-7617.
- 39 Govindasamy, B., P.B. Duffy, and K. Caldeira, 2001a: Land use changes and Northern hemisphere cooling.  
40 *Geophys. Res. Lett.*, **28**(2), 291-294.
- 41 Govindasamy, B., et al., 2001b: Limitations of the equivalent CO<sub>2</sub> approximation in climate change  
42 simulations. *J. Geophys. Res.*, **106**(D19), 22593-22603.
- 43 Gray, L.J., J.D. Haigh, and R.G. Harrison, 2005: *Review of the influences of solar changes on the Earth's*  
44 *climate*. Hadley Centre Technical Note No. 62, Met Office, Exeter, 82 p.
- 45 Gregory, J.M., et al., 2004: A new method for diagnosing radiative forcing and climate sensitivity. *Geophys.*  
46 *Res. Lett.*, **31**, L03205, doi:10.1029/2003GL018747.
- 47 Gunn R, and B.B. Phillips, 1957: An experimental investigation of the effect of air pollution on the initiation  
48 of rain. *J. Meteor.* **14**(3), 272-280.
- 49 Hadjinicolaou, P., J.A. Pyle, and N.R.P. Harris, 2005: The recent turnaround in stratospheric ozone over  
50 northern middle latitudes: a dynamical modeling perspective. *Geophys. Res. Lett.*, **32**, L12821,  
51 doi:10.1029/2005GL022476.
- 52 Haigh, J.D., 2003: The effects of solar variability on the Earth's climate. *Phil. Trans. R. Soc.*, **361A**, 95-111.
- 53 Hall, J.C., and G.W. Lockwood, 2004: The chromospheric activity and variability of cycling and flat activity  
54 solar-analog stars. *Astrophys. J.*, **614**, 942-946.
- 55 Hamilton, J.F., et al., 2004: Partially oxidised organic components in urban aerosol using GCXGC-TOF/MS.  
56 *Atmos. Chem. Physics*, **4**, 1279-1290.

- 1 Hammitt, J.K., A.K. Jain, J.L. Adams, and D.J. Wuebbles, 1996: A welfare-based index for assessing  
2 environmental effects of greenhouse-gas emissions. *Nature*, **381**, 301-303.
- 3 Han, Q., W.B. Rossow, J. Zeng, and R. Welch, 2002: Three different behaviors of liquid water path of water  
4 clouds in aerosol-cloud interactions. *J. Atmos. Sci.*, **59**, 726-735.
- 5 Hansen, J., and L. Nazarenko, 2004: Soot climate forcing via snow and ice albedos. *Proc. Natl. Acad. Sci.*  
6 *U.S.A.*, **101**, 423-428.
- 7 Hansen, J., and M. Sato, 2001: Trends of measured climate forcing agents. *Proc. Natl. Acad. Sci. U.S.A.*, **98**,  
8 14778-14783.
- 9 Hansen, J., and M. Sato, 2004: Greenhouse gas growth rates. *Proc. Natl. Acad. Sci. U.S.A.*, **101**, 16109-  
10 16114.
- 11 Hansen, J., M. Sato, R. Ruedy, 1995: Long-term changes of the diurnal temperature cycle: Implications  
12 about mechanisms of global climate change. *Atmos. Res.*, **37**, 175-209.
- 13 Hansen, J., M. Sato, and R. Ruedy, 1997: Radiative forcing and climate response. *J. Geophys. Res.*, **102**(D6),  
14 6831-6864.
- 15 Hansen, J., M. Sato, R. Ruedy, A. Lacis, and V. Oinas, 2000: Global warming in the twenty-first century: An  
16 alternative scenario. *Proc. Natl. Acad. Sci. U.S.A.*, **97**, 9875-9880.
- 17 Hansen, J., et al., 2002: Climate forcings in Goddard Institute for Space Studies SI2000 simulations. *J.*  
18 *Geophys. Res.*, **107**(D18), 4347,10.1029/2001JD001143.
- 19 Hansen, J., et al., 2005: Efficacy of climate forcings. *J. Geophys. Res.*, **110**, D18104,  
20 doi:10.1029/2005JD005776.
- 21 Hansen, M.C., and B. Reed, 2000: A comparison of the IGBP DISCover and University of Maryland 1km  
22 global land cover products. *Int. J. Remote Sensing*, **21**, 1365-1373.
- 23 Harder, J., G. Lawrence, J. Fontenla, G. Rottman, and T. Woods, 2005: The spectral irradiance monitor I.  
24 Scientific requirements, instrument design and operation modes. *Sol. Phys.*, **230**, 141-167.
- 25 Harding, R.J., and J.W. Pomeroy, 1996: The energy balance of the winter boreal landscape. *J. Climate*, **9**,  
26 2778-2787.
- 27 Harnisch, J., R.R. Borchers, P.P. Fabian, and M.M. Maiss, 1996: Tropospheric trends for CF4 and CF3CF3  
28 since 1982 derived from SF6 dated stratospheric air. *Geophys. Res. Lett.*, **23**(10), 1099-1102.
- 29 Harries, J.E., H.E. Brindley, P.J. Sagoo, and R.J. Bantges, 2001: Increases in greenhouse forcing inferred  
30 from the outgoing longwave radiation spectra of the Earth in 1970 and 1997. *Nature*, **410**, 355-357.
- 31 Harrison, R.G., and K.S. Carslaw, 2003: Ion-aerosol-cloud processes in the lower atmosphere. *Rev.*  
32 *Geophys.*, **41**, 1021.
- 33 Harrison R.G., and D.B. Stephenson, 2006: Empirical evidence for a nonlinear effect of galactic cosmic rays  
34 on clouds. *Proc. Roy. Soc. A*, **462**, 1221-1233.
- 35 Hartmann, D.L., M.E. Ockert-Bell, and M.L. Michelsen, 1992: The effect of cloud type on Earth's energy  
36 balance: global analysis. *J. Climate*, **5**, 1281-1304.
- 37 Hashvardhan, S.E. Schwartz, C.E. Benkovitz, and G. Guo, 2002: Aerosol influence on cloud microphysics  
38 examined by satellite measurements and chemical transport modelling. *J. Atmos. Sci.*, **59**, 714-725.
- 39 Hauglustaine, D.A., and G.P. Brasseur, 2001: Evolution of tropospheric ozone under anthropogenic activities  
40 and associated radiative forcing of climate. *J. Geophys. Res.*, **106**(D23), 32337-32360,  
41 10.1029/2001JD900175.
- 42 Haxeltine, A. and I.C. Prentice, 1996: BIOME3: An equilibrium terrestrial biosphere model based on  
43 ecophysiological constraints, resource availability, and competition among plant functional types.  
44 *Global Biogeochem. Cycles*, **10**(4), 693-709.
- 45 Haywood, J., and O. Boucher, 2000: Estimates of the direct and indirect radiative forcing due to tropospheric  
46 aerosols: A review. *Rev. Geophys.*, **38**, 513-543.
- 47 Haywood, J.M., and K.P. Shine, 1995: The effect of anthropogenic sulfate and soot aerosol on the clear sky  
48 planetary radiation budget. *Geophys. Res. Lett.*, **22**(5), 603-606.
- 49 Haywood, J.M., and K.P. Shine. 1997: Multi-spectral calculations of the direct radiative forcing of  
50 tropospheric sulphate and soot aerosols using a column model. *Q. J.R. Meteorol. Soc.* **123**:1907-  
51 1930.
- 52 Haywood, J.M., V. Ramaswamy, and B.J. Soden, 1999: Tropospheric aerosol climate forcing in clear-sky  
53 satellite observations over the oceans. *Science*, **283**, 1299-1305.
- 54 Haywood, J.M., P. Francis, O. Dubovik, M. Glew, and B. Holben, 2003a: Comparison of aerosol size  
55 distributions, radiative properties, and optical depths determined by aircraft observations and Sun  
56 photometers during SAFARI 2000. *J. Geophys. Res.*, **108**(D13), 8471, doi:10.1029/2002JD002250.

- 1 Haywood, J.M., et al., 2003b: The mean physical and optical properties of regional haze dominated by  
2 biomass burning aerosol measured from the C-130 aircraft during SAFARI 2000. *J. Geophys. Res.*,  
3 **108**(D13), 8473, doi:10.1029/2002JD002226.
- 4 Haywood, J.M., et al., 2003c: Radiative properties and direct radiative effect of Saharan dust measured by the  
5 C-130 aircraft during SHADE: 1. Solar spectrum. *J. Geophys. Res.*, **108**(D18), 8577,  
6 doi:10.1029/2002JD002687.
- 7 Haywood, J.M., et al., 2005: Can desert dust explain the outgoing longwave radiation anomaly over the  
8 Sahara during July 2003? *J. Geophys. Res.*, **110**, D05105, doi:10.1029/2004JD005232.
- 9 Heal, G., 1997: Discounting and climate change: an editorial comment. *Clim. Change*, **37**, 335-343.
- 10 Hegg, D.A., J. Livingston, P.V. Hobbs, T. Novakov, and P. Russell, 1997: Chemical apportionment of  
11 aerosol column optical depth off the mid-Atlantic coast of the United States. *J. Geophys. Res.*,  
12 **102**(D21), 25,293-25,303.
- 13 Hély, C., K. Caylor, S. Alleaume, R.J. Swap, and H.H. Shugart, 2003: Release of gaseous and particulate  
14 carbonaceous compound from biomass burning during the SAFARI 2000 dry season field campaign.  
15 *J. Geophys. Res.*, **108**(D13), 8470, doi:10.1029/2002JD002482.
- 16 Hendricks, J., B. Kärcher, U. Lohmann, and M. Ponater, 2005: Do aircraft black carbon emissions affect  
17 cirrus clouds on the global scale? *Geophys. Res. Lett.*, **32**, L12814, doi: 10.1029/2005GL022740.
- 18 Hendricks, J., et al., 2004: Simulating the global atmospheric black carbon cycle: a revisit to the contribution  
19 of aircraft. *Atmos. Chem. Phys.*, **4**, 2521-2541.
- 20 Herman, M., et al. 1997: Remote sensing of aerosols over land surfaces including polarization measurements  
21 and application to POLDER measurements. *J. Geophys. Res.*, **102**(D14), 17039-17050,  
22 10.1029/96JD02109.
- 23 Hess, M., P. Koepke, and I. Schult, 1998: Optical properties of aerosols and clouds: the software package  
24 OPAC. *Bull. Am. Meteorol. Soc.*, **79**, 831-844.
- 25 Highwood, E.J., J.M. Haywood, M.D. Silverstone, S.M. Newman, and J.P. Taylor, 2003: Radiative  
26 properties and direct effect of Saharan dust measured by the C-130 aircraft during Saharan Dust  
27 Experiment (SHADE): 2. Terrestrial spectrum. *J. Geophys. Res.*, **108**(D18), 8577,  
28 doi:10.1029/2002JD002552.
- 29 Higurashi, A., and T. Nakajima. 1999. Development of a two-channel aerosol retrieval algorithm on a global  
30 scale using NOAA AVHRR. *J. Atmos. Sci.* **56**, 924-941.
- 31 Higurashi, A., et al., 2000: A study of global aerosol optical climatology with two-channel AVHRR remote  
32 sensing. *J. Climate*, **13**, 2011-2027.
- 33 Hirsch, A. I., et al., 2006: Inverse modeling estimates of the global nitrous oxide surface flux from 1998–  
34 2001. *Global Biogeochem. Cycles*, **20**, GB1008, doi:10.1029/2004GB002443.
- 35 Hirst, E., P.H. Kaye, R.S. Greenaway, P.R. Field, and D.W. Johnson, 2001: Discrimination of micrometre-  
36 sized ice and super-cooled droplets in mixed-phase cloud. *Atmos. Environ.*, **35**, 33-47.
- 37 Hofmann, D., 2003: et al., Surface-based observations of volcanic emissions to stratosphere. In: *Volcanism  
38 and the Earth's Atmosphere*[Robock, A. and C. Oppenheimer, (eds.)]. Geophysical Monograph 139,  
39 American Geophysical Union, Washington, DC, pp. 57-73.
- 40 Holben, B.N., et al., 1998: AERONET: A federated instrument network and data archive for aerosol  
41 characterization. *Remote Sens. Environ.*, **66**, 1-16.
- 42 Holben, B.N., et al., 2001: An emerging ground-based aerosol climatology: aerosol optical depth from  
43 AERONET. *J. Geophys. Res.*, **106**(D11), 12067-12097.
- 44 Holzer-Popp, T., M. Schroedter, and G. Gesell, 2002: Retrieving aerosol optical depth and type in the  
45 boundary layer over land and ocean from simultaneous GOME spectrometer and ATSR-2 radiometer  
46 measurements - 2. Case study application and validation. *J. Geophys. Res.*, **107**(D21), 4578,  
47 doi:10.1029/2002JD002777.
- 48 Hood, L.L., 2003: Thermal response of the tropical tropopause region to solar ultraviolet variations.  
49 *Geophys. Res. Lett.*, **30**, 2215, doi:10.1029/2003GL018364.
- 50 Houghton, R.A., et al., 1983: Changes in the carbon content of terrestrial biota and soils between 1860 and  
51 1980: a net release of CO<sub>2</sub> to the atmosphere. *Ecol. Monogr.*, **53**, 235-262, doi:10.2307/1942531
- 52 Houghton, R.A. 1999: The annual net flux of carbon to the atmosphere from changes in land use 1850-1990.  
53 *Tellus*, **51B**, 298-313
- 54 Houghton, R.A. 2000: A new estimate of global sources and sinks of carbon from land use change. *EOS* **81**  
55 (19, Supplement), S281
- 56 Houghton, R.A., 2003: Revised estimates of the annual net flux of carbon to the atmosphere from changes in  
57 land use and land management. *Tellus*, **55B**, 378-390.

- 1 von Hoyningen-Huene, W., M. Freitag, and J.B. Burrows, 2003: Retrieval of aerosol optical thickness over  
2 land surfaces from top-of-atmosphere radiance. *J. Geophys. Res.*, **108**(D9), 4260,  
3 doi:10.1029/2001JD002018.
- 4 Hoyt, D.V., and K.H. Schatten, 1993: A discussion of plausible solar irradiance variations, 1700-1992. *J.*  
5 *Geophys. Res.*, **98**(A11), 18895-18906.
- 6 Hsu, N.C., J.R. Herman, and C. Weaver, 2000: Determination of radiative forcing of Saharan dust using  
7 combined TOMS and ERBE data. *J. Geophys. Res.*, **105**(D16), 20,649-20,661.
- 8 Hsu, N.C., J.R. Herman, and S.C. Tsay, 2003: Radiative impacts from biomass burning in the presence of  
9 clouds during boreal spring in southeast Asia. *Geophys. Res. Lett.*, **30**(5), 28,  
10 doi:10.1029/2002GL016485.
- 11 Huang, J., and R.G. Prinn, 2002: Critical evaluation of emissions of potential new gases for OH estimation.  
12 *J. Geophys. Res.*, **107**(D24), 4784, doi:10.1029/2002JD002394.
- 13 Huebert, B.J., et al., 2003: An overview of ACE-Asia: Strategies for quantifying the relationships between  
14 Asian aerosols and their climatic impacts. *J. Geophys. Res.*, **108**(D23), 8633,  
15 doi:10.1029/2003JD003550.
- 16 Hurley, M.D., et al., 2005: IR spectrum and radiative forcing of CF<sub>4</sub> revisited. *J. Geophys. Res.*, **110**,  
17 D02102, doi:10.1029/2004JD005201.
- 18 Hurst, D.F., et al., 2004: Emissions of ozone-depleting substances in Russia during 2001. *J. Geophys. Res.*,  
19 **109**, D14303, doi:10.1029/2004JD004633.
- 20 Husar, R.B., J.M. Prospero, and L.L. Stowe, 1997: Characterization of tropospheric aerosols over the oceans  
21 with the NOAA advanced very high resolution radiometer optical thickness operational product. *J.*  
22 *Geophys. Res.*, **102**(D14), 16889-16910.
- 23 Husar, R.B., et al., 2001: Asian dust events of April 1998. *J. Geophys. Res.*, **106**(D16), 18317-18330.
- 24 Ichinose, T., K. Shimodozono, and K. Hanaki, 1999: Impact of anthropogenic heat on urban climate in  
25 Tokyo. *Atmos. Env.*, **33**, 3897-3909.
- 26 Ichoku, C., et al., 2003: MODIS observation of aerosols and estimation of aerosol radiative forcing over  
27 southern Africa during SAFARI 2000. *J. Geophys. Res.*, **108**(D13), 8499,  
28 doi:10.1029/2002JD002366.
- 29 Ignatov, A., and L. Stowe, 2002: Aerosol retrievals from individual AVHRR channels. Part I: Retrieval  
30 algorithm and transition from Dave to 6S Radiative Transfer Model. *J. Atmos. Sci.*, **59**, 313-334.
- 31 IPCC, 1990: *Climate Change: The Intergovernmental Panel on Climate Change Scientific Assessment*  
32 [Houghton, J.T., G.J. Jenkins, and J.J. Ephraums (eds.)], Cambridge University Press, Cambridge,  
33 UK, 364 p.
- 34 IPCC, 1999: *Aviation and the global atmosphere: a special report of the IPCC Working Group I and III*  
35 [Penner, J.E., D.H. Lister, D.J. Griggs, D.J. Dokken, and M. MacFarland (eds.)]. Cambridge  
36 University Press, 373 p.
- 37 IPCC, 2001: *Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third*  
38 *Assessment Report of the Intergovernmental Panel on Climate Change (IPCC)* [Houghton, J.T., et al.  
39 (eds.)]. Cambridge University Press, Cambridge, UK, 881 p..
- 40 IPCC/TEAP, 2005: *Special Report on Safeguarding the Ozone Layer and the Global Climate System: Issues*  
41 *Related to Hydrofluorocarbons and Perfluorocarbons* [Metz, B., et al. (eds.)]. Cambridge University  
42 Press, Cambridge, UK, 488 p..
- 43 Ito, A., and J. E. Penner, 2005: Historical emissions of carbonaceous aerosols from biomass and fossil fuel  
44 burning for the period 1870–2000. *Global Biogeochem. Cycles*, **19**, GB2028,  
45 doi:10.1029/2004GB002374.
- 46 Iversen, T., and O. Seland, 2002: A scheme for process-tagged SO<sub>4</sub> and BC aerosols in NCAR CCM3:  
47 Validation and sensitivity to cloud processes. *J. Geophys. Res.*, **107**, (D24), 4751,  
48 doi:10.1029/2001JD000885.
- 49 Jacob, D.J., et al., 2005: *Radiative forcing of climate change*. The National Academies Press, Washington,  
50 D.C., 207 p.
- 51 Jacobson, M.Z., 1999: Isolating nitrated and aromatic aerosols and nitrated aromatic gases as sources of  
52 ultraviolet light absorption. *J. Geophys. Res.*, **104**(D3), 3527-3542.
- 53 Jacobson, M.Z., 2001a: Global direct radiative forcing due to multicomponent anthropogenic and natural  
54 aerosols. *J. Geophys. Res.*, **106**(D2), 1551-1568.
- 55 Jacobson, M.Z., 2001b: Strong radiative heating due to the mixing state of black carbon in atmospheric  
56 aerosols. *Nature*, **409**, 695-697.

- 1 Jacobson, M.Z., 2002: Control of fossil-fuel particulate black carbon and organic matter, possibly the most  
2 effective method of slowing global warming. *J. Geophys. Res.*, **107**(D19), 4410, doi:  
3 10.1029/2001JD001376.
- 4 Jacobson, M.Z., 2004: Climate response of fossil fuel and biofuel soot, accounting for soot's feedback to  
5 snow and sea ice albedo and emissivity. *J. Geophys. Res.*, **109**, D21201, doi:10.1029/2004JD004945.
- 6 Jaffe, D., H.Price, D.D. Parrish, A.H. Goldstein, and J. Harris, 2003: Increasing background ozone during  
7 spring on the west coast of North America. *Geophys. Res. Lett.*, **30**, 1613,  
8 doi:10.1029/2003GL017024.
- 9 Jensen, E.J., and O.B. Toon, 1997: The potential impact of soot particles from aircraft exhaust on cirrus  
10 clouds. *Geophys. Res. Lett.*, **24**(3), 249-252.
- 11 Jeong, M.J., Z.Q. Li, D.A. Chu, and S.C. Tsay, 2005: Quality and compatibility analyses of global aerosol  
12 products derived from the advanced very high resolution radiometer and Moderate Resolution  
13 Imaging Spectroradiometer. *J. Geophys. Res.*, **110**. D10S09 doi:10.1029/2004JD004648.
- 14 Jiang, H., G. Feingold, and W.R. Cotton, 2002: Simulations of aerosol-cloud-dynamical feedbacks resulting  
15 from of entrainment of aerosols into the marine boundary layer during the Atlantic Stratocumulus  
16 Transition Experiment. *J. Geophys. Res.*, **107**(D24), 4813, ,doi:10.1029/2001JD001502.
- 17 Jiang, H. and G. Feingold, 2006: Effect of aerosol on warm convective clouds: Aerosol-clouds-  
18 surface flux feedbacks in a new coupled large eddy model. *J. Geophys. Res.*, **111**, D01202,  
19 doi:10.1029/2005JD006138.
- 20 Jirikowic, J.L., and P.E. Damon, 1994: The Medieval solar activity maximum. *Clim. Change*, **26**, 309-316.
- 21 Johnson, B.T., 2005: The semidirect aerosol effect: comparison of a single-column model with large eddy  
22 simulation for marine stratocumulus. *J. Climate*, **18**, 119-130.
- 23 Johnson, B.T., K.P. Shine, and P.M. Forster, 2004: The semi-direct aerosol effect: Impact of absorbing  
24 aerosols on marine stratocumulus. *Q.J.R. Meteorol. Soc.*, **130**, 1407-1422.
- 25 Jones, A., D.L. Roberts, and M.J. Woodage, 2001: Indirect sulphate aerosol forcing in a climate model with  
26 an interactive sulphur cycle. *J. Geophys. Res.*, **106**(D17), 20293-20301.
- 27 Jones, P.D., A. Moberg, T.J. Osborn, and K.R. Briffa, 2003: Surface Climate Responses to Explosive  
28 Volcanic Eruptions Seen in Long European Temperature Records and Mid-to-High Latitude Tree-  
29 Ring Density Around the Northern Hemisphere. In: *Volcanism and the Earth's Atmosphere* [Rohock,  
30 A. and C. Oppenheimer (eds.)]Geophysical Monograph 139, American Geophysical Union, pp. 239-  
31 254.
- 32 Jonson, J. E., D. Simpson, H. Fagerli, and S. Solberg, 2005: Can we explain the trends in European ozone  
33 levels? *Atmos. Chem. Phys. Disc.*, **5**, 5957-5985.
- 34 Joos, F. et al. 1996. An efficient and accurate representation of complex oceanic and biospheric models of  
35 anthropogenic carbon uptake. *Tellus*, 48B, 397-417.
- 36 Joshi, M., et al., 2003: A comparison of climate response to different radiative forcings in three general  
37 circulation models: Towards an improved metric of climate change. *Clim. Dyn.*, **20**, 843-854.
- 38 Joshi, M.M., and K.P. Shine, 2003: A GCM study of volcanic eruptions as a cause of increased stratospheric  
39 water vapor. *J. Climate*, **16**, 3525-3534.
- 40 Junge, C.E., 1975: The possible influence of aerosols on the general circulation and climate and possible  
41 approaches for modelling. In: *The physical basis of climate and climate modelling : report of the*  
42 *International Study Conference in Stockholm, 29 July-10 August 1974 : organised by WMO and*  
43 *ICSU and supported bu UNEP Global Atmospheric Research Programme (GARP), WMO-ICSU*  
44 *Joint Organising Committee*. WMO ICSU, Geneva, pp. 244-251.
- 45 Kahn, R., P. Banerjee, and D. McDonald, 2001: Sensitivity of multiangle imaging to natural mixtures of  
46 aerosols over ocean. *J. Geophys. Res.*, **106**(D16), 18219-18238.
- 47 Kahn, R.A., et al., 2005: Multiangle Imaging Spectroradiometer (MISR) global aerosol optical depth  
48 validation based on 2 years of coincident Aerosol Robotic Network (AERONET) observation. *J.*  
49 *Geophys. Res.*, **110**, D10S04, doi:10.1029/2004JD004706.
- 50 Kalkstein, A.J., and R.C. Balling Jr., 2004: Impact of unusually clear weather on United States daily  
51 temperature range following 9/11/2001. *Clim. Res.*, **26**, 1-4.
- 52 Kanakidou, M. et al., 2005: Organic aerosol and global climate modelling: a review. *Atmos. Chem. Phys.*, **5**,  
53 1053-1123.
- 54 Kandlikar, M., 1996: Indices for comparing greenhouse gas emissions: integrating science and economics.  
55 *Energ. Econ.*, **18**, 265-282.
- 56 Kapustin V.N., et al., 2006: On the determination of a cloud condensation nuclei from satellite: challenges and  
57 possibilities. *J. Geophys. Res.*, **111**, D04202, doi:10.1029/2004JD005527.

- 1 Kärcher, B., 1999: Aviation-produced aerosols and contrails. *Surv. Geophys.*, **20**, 113-167.
- 2 Kasischke, E.S. and J.E. Penner, 2004: Improving global estimates of atmospheric emissions from biomass  
3 burning. *J. Geophys. Res.*, **109**, D14S01, doi:10.1029/2004JD004972.
- 4 Kaufman, Y.J., C.J. Tucker and R.L. Mahoney, 1991: Fossil fuel and biomass burning effect on climate:  
5 Heating or cooling? *J. Climate*, **4**, 578-588.
- 6 Kaufman, Y.J., D. Tanré, and O. Boucher, 2002: A satellite view of aerosols in the climate system. *Nature*,  
7 **419**, 215-223.
- 8 Kaufman, Y.J., D. Tanré, L. Remer, E. Vermote, and B.N. Holben, 1997: Operational remote sensing of  
9 tropospheric aerosols over the land from EOS-MODIS. *J. Geophys. Res.*, **102**(D14), 17051-17068.
- 10 Kaufman, Y.J., D. Tam, O. Dubovik, A. Karnieli, and L.A. Remer, 2001: Absorption of sunlight by dust as  
11 inferred from satellite and ground-based remote sensing. *Geophys. Res. Lett.*, **28**(8), 1479-1482.
- 12 Kaufman Y.J., et al., 2005a: Aerosol anthropogenic component estimated from satellite data. *Geophys. Res.*  
13 *Lett.*, **32**, L17804, doi:10.1029/2005GL023125.
- 14 Kaufman, Y.J., et al., 2005b: A critical examination of the residual cloud contamination and diurnal  
15 sampling effects on MODIS estimates of aerosol over ocean. *IEEE T. Geosci. Remote*, **43** (12),  
16 2886-2897
- 17 Keeling, C.D., and T.P. Whorf, 2005: Atmospheric CO<sub>2</sub> records from sites in the SIO air sampling network.  
18 In: *Trends: A Compendium of Data on Global Change*. Carbon Dioxide Information Analysis  
19 Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tenn., U.S.A.  
20 <http://cdiac.esd.ornl.gov/trends/co2/sio-keel-flask/sio-keel-flask.html>
- 21 Keeling, C.D., T.P. Whorf, M. Wahlen, and J. van der Plicht, 1995: Interannual extremes in the rate of rise of  
22 atmospheric carbon dioxide since 1980. *Nature*, **375**, 666-670.
- 23 Keeling, C.D., A.F. Bollenbacher, and T.P. Whorf, 2005: Monthly atmospheric <sup>13</sup>C/<sup>12</sup>C isotopic ratios for 10  
24 SIO stations. In: *Trends: A Compendium of Data on Global Change*. Carbon Dioxide Information  
25 Analysis Center, Publication No. ORNL/CDIAC-65, Oak Ridge National Laboratory, U.S.  
26 Department of Energy, Oak Ridge, Tenn., U.S.A. <http://cdiac.ornl.gov/trends/co2/iso-sio/iso-sio.html>  
27
- 28 Keeling, R.F., and S.R. Shertz, 1992: Seasonal and interannual variations in atmospheric oxygen and  
29 implications for the global carbon cycle. *Nature*, **358**, 723-727.
- 30 Keeling, R.F., S.C. Piper, and M. Heimann, 1996: Global and hemispheric CO<sub>2</sub> sinks deduced from changes  
31 in atmospheric O<sub>2</sub> concentration. *Nature*, **381**, 218-221.
- 32 Keil, A., and J.M. Haywood, 2003: Solar radiative forcing by biomass burning aerosol particles during  
33 SAFARI 2000: A case study based on measured aerosol and cloud properties. *J. Geophys. Res.*, **108**  
34 (D13), 8467, doi:10.1029/2002JD002315.
- 35 Keppler, F., J.T.G. Hamilton, M. Brass, T. Röckmann, 2006: Methane emissions from terrestrial plants under  
36 aerobic conditions. *Nature*, **439**, 187-191, doi :10.1038/nature04420.
- 37 Kernthaler, S.C., R. Toumi, J.D. Haigh, 1999: Some doubts concerning a link between cosmic ray fluxes and  
38 global cloudiness, *Geophys. Res. Lett.*, **26**(7), 863-866, 10.1029/1999GL900121.
- 39 Khainroutdinov, M., and Y. Kogan, 2000: A new cloud physics parameterization in a large-eddy simulation  
40 model of marine stratocumulus. *Mon. Wea. Rev.*, **128**, 229-243.
- 41 Khalil, M.A.K., et al., 2003: Atmospheric perfluorocarbons. *Environ. Sci. Technol.*, **37**, 4358-4361.
- 42 Kiehl, J.T., T.L. Schneider, P.J. Rasch, M.C. Barth, and J. Wong, 2000: Radiative forcing due to sulfate  
43 aerosols from simulations with the National Center for Atmospheric Research Community Climate  
44 Model, Version 3. *J. Geophys. Res.*, **105**(D1), 1441-1457.
- 45 Kim, B.-G., S.E. Schwartz, and M.A. Miller, 2003: Effective radius of cloud droplets by ground-based  
46 remote sensing: Relationship to aerosol. *J. Geophys. Res.*, **108**(D23), doi: 10.1029/2003JD003721.
- 47 Kinne, S., et al., 2006: An AeroCom initial assessment: optical properties in aerosol component modules of  
48 global models. *Atmos. Chem. Phys.* **6**, 1815-1834.
- 49 Kinne, S., et al., 2003: Monthly averages of aerosol properties: a global comparison among models, satellite  
50 data, and AERONET ground data. *J. Geophys. Res.*, **108** (D20), 4634, doi:10.1029/2001JD001253.
- 51 Kirchstetter, T.W., T. Novakov, and P.V. Hobbs, 2004: Evidence that the spectral dependence of light  
52 absorption by aerosols is affected by organic carbon. *J. Geophys. Res.*, **109**, D21208,  
53 doi:10.1029/2004JD004999.
- 54 Kirkevåg, A., and T. Iversen, 2002: Global direct radiative forcing by process-parameterized aerosol optical  
55 properties. *J. Geophys. Res.*, **107**(D20), 4433, doi:10.1029/2001JD000886.
- 56 Klein Goldewijk, K., 2001: Estimating global land use change over the past 300 years: The HYDE database.  
57 *Global Biogeochem. Cycles*, **15**, 417-433.

- 1 Knutson, T. R., et al., 2006: Assessment of twentieth-century regional surface temperature trends using the  
2 GFDL CM2 coupled models. *J. Climate*, **10**(9), 1624-1651.
- 3 Koch, D., 2001: Transport and direct radiative forcing of carbonaceous and sulfate aerosols in the GISS  
4 GCM. *J. Geophys. Res.*, **106**(D17), 20311-20332.
- 5 Korolev, A.V. and G.A. Isaac, 2005: Shattering during sampling OAPs and HVPS. Part I: snow particles. *J.*  
6 *Atmos. and Ocean. Technol.*, **22**, 528-542.
- 7 Kotchenruther, R.A., and P.V. Hobbs, 1998: Humidification factors of aerosols from biomass burning in  
8 Brazil. *J. Geophys. Res.*, **103**(D24), 32081-32089.
- 9 Kotchenruther, R.A., P.V. Hobbs, and D.A. Hegg, 1999: Humidification factors for atmospheric aerosols off  
10 the mid-Atlantic coast of the United States. *J. Geophys. Res.*, **104**(D2), 2239-2251.
- 11 Kristjánsson, J.E. and J. Kristiansen, 2000: Is there a cosmic ray signal in recent variations in global  
12 cloudiness and cloud radiative forcing? *J. Geophys. Res.*, **105**(D9), 11851-11863.
- 13 Kristjánsson, J.E., A. Staple, J. Kristiansen, and E. Kaas, 2002: A new look at possible connections between  
14 solar activity, clouds and climate. *Geophys. Res. Lett.*, **29**, doi:10.1029/2002GL015646.
- 15 Kristjánsson, E., T. Iversen, A. Kirkevåg, Ø. Seland and J. Debernard, 2005: Response of the climate system  
16 to aerosol direct and indirect forcing: the role of cloud feedbacks. *J. Geophys. Res.*, **110**, D24206,  
17 doi:10.1029/2005JD006299
- 18 Krizan P., J. Lastovicka, 2005: Trends in positive and negative ozone laminae in the Northern Hemisphere.  
19 *J. Geophys. Res.*, **110**, D10107, doi:10.1029/2004JD005477
- 20 Kroeze, C., E. Dumont, and S.P. Seitzinger, 2005: New estimates of global emissions of N<sub>2</sub>O from rivers,  
21 estuaries and continental shelves. In: *Non-CO<sub>2</sub> Greenhouse Gases* [van Amstel, A., and T. Pulles  
22 (eds.)] (published as Environmental Sciences Vol. 2, No. 2-3, Special Issue), pp. 159-165.
- 23 Krol, M., and J. Lelieveld, 2003: Can the variability in tropospheric OH be deduced from measurements of  
24 1,1,1-trichloroethane (methyl chloroform)? *J. Geophys. Res.*, **108**(D3), 4125,  
25 doi:10.1029/2002JD002423.
- 26 Krol, M., et al., 2003: Continuing emissions of methyl chloroform from Europe. *Nature*, **421**, 131-135.
- 27 Krüger, O. and H. Graßl, 2002: The indirect aerosol effect over Europe. *Geophys. Res. Lett.*, **29**,  
28 doi:10.1029/2001GL14081.
- 29 Krueger A.J., S.J. Schaefer, N.A. Krotkov, G. Bluth, and S. Barker, 2000: Ultraviolet remote sensing of  
30 volcanic emissions. In: *Remote Sensing of Active Volcanism* [Mouginis-Mark, P.J., J.A. Crisp, and  
31 J.H. Fink, (eds.)]. Geophysical Monograph 116, American Geophysical Union, Washington, D.C.,  
32 pp. 25-43.
- 33 Labitzke, K., 2004: On the signal of the 11-year sunspot cycle in the stratosphere and its modulation by the  
34 quasi-biennial oscillation. *J. Atmos. Solar Terr. Phys.*, **66**, 1151-1157.
- 35 Labitzke, K., et al., 2002: The global signal of the 11-year solar cycle in the stratosphere: observations and  
36 models. *J. Atmos. Solar Terr. Phys.*, **64**, 203-210.
- 37 Lambert, A., et al., 1993: Measurements of the evolution of the Mt. Pinatubo aerosol cloud by ISAMS.  
38 *Geophys. Res. Lett.*, **20**(12), 1287-1290
- 39 Langenfelds, R.L., et al., 2002: Interannual growth rate variations of atmospheric CO<sub>2</sub> and its delta C-13, H-  
40 2, CH<sub>4</sub>, and CO between 1992 and 1999 linked to biomass burning. *Global Biogeochem. Cycles*, **16**,  
41 doi: 10.1029/2001GB001466.
- 42 Lawrence, M.G., P. Jöckel, and R. von Kuhlmann, 2001: What does the global mean OH concentration tell  
43 us? *Atmos. Chem. Phys.*, **1**, 43-74.
- 44 Lean, J., 2000: Evolution of the sun's spectral irradiance since the Maunder Minimum. *Geophys. Res. Lett.*,  
45 **27**, 2425-2428.
- 46 Lean, J., A. Skumanich, and O. White, 1992: Estimating the sun's radiative output during the Maunder  
47 Minimum. *Geophys. Res. Lett.*, **19**(15), 1595-1598.
- 48 Lean, J., J. Beer, and R. Bradley, 1995: Reconstruction of solar irradiance since 1610: implications for  
49 climate change. *Geophys. Res. Lett.*, **22**, 3195-3198.
- 50 Lean, J.L., et al. 1997: Detection and parameterization of variations in solar mid and near ultraviolet radiation  
51 (200 to 400 nm). *J. Geophys. Res.*, **102**(D25), 29939-29956.
- 52 Lean, J.L., Y.M. Wang, and N.R. Sheeley, 2002: The effect of increasing solar activity on the Sun's total and  
53 open magnetic flux during multiple cycles: Implications for solar forcing of climate. *Geophys. Res.*  
54 *Lett.*, **29**(24), 2224, doi:10.1029/2002GL015880.
- 55 Lean, J., G. Rottman, J. Harder, and G. Kopp, 2005: SORCE contribution to new understanding of global  
56 change and solar variability. *Solar Phys.*, **230**, 27-53.

- 1 Lee, R.B., III, M.A. Gibson, R.S. Wilson, and S. Thomas, 1995: Long-term total solar irradiance variability  
2 during sunspot cycle 22. *J. Geophys. Res.*, **100**(A2), 1667-1675.
- 3 Lee, K.H., Y.J. Kim, and W. von Hoyningen-Huene, 2004: Estimation of regional aerosol optical thickness  
4 from satellite observations during the 2001 ACE-Asia IOP. *J. Geophys. Res.* **109**, D19S16,  
5 doi:10.1029/2003JD004126.
- 6 Lefohn, A.S., J.D. Husar, and R.B. Husar, 1999: Estimating historical anthropogenic global sulfur emission  
7 patterns for the period 1850-1990. *Atmos. Env.*, **33**, 3435-3444.
- 8 Lelieveld, J., P.J. Crutzen and F.J. Dentener, 1998: Changing concentration, lifetime and climate forcing of  
9 atmospheric methane. *Tellus* **50B**, 128-150.
- 10 Lelieveld, J., W. Peters, F.J. Dentener and M.C. Krol, 2002: Stability of tropospheric hydroxyl chemistry. *J.*  
11 *Geophys. Res.* **107**(D23), 4715, doi: 10.1029/2002JD002272.
- 12 Lelieveld, J., et al., 2004: Increasing ozone over the Atlantic Ocean. *Science*, **304**, 1483-1487.
- 13 Levy R. C., et al., 2003: Evaluation of the Moderate-Resolution Imaging Spectroradiometer (MODIS)  
14 retrievals of dust aerosol over the ocean during PRIDE. *J. Geophys. Res.*, **108** (D19), 8594,  
15 doi:10.1029/2002JD002460.
- 16 Li, J., et al., 2005: Halocarbon emissions estimated from AGAGE measured pollution events at Trinidad  
17 Head, California. *J. Geophys. Res.*, **110**, D14308, doi:10.1029/2004JD005739.
- 18 Liao, H., and J.H. Seinfeld, 2005: Global impacts of gas-phase chemistry-aerosol interactions on direct  
19 radiative forcing by anthropogenic aerosols and ozone. *J. Geophys. Res.*, **110**, D18208,  
20 doi:10.1029/2005JD005907.
- 21 Liao, H., J.H. Seinfeld, P.J. Adams, and L.J. Mickley, 2004: Global radiative forcing of coupled tropospheric  
22 ozone and aerosols in a unified general circulation model. *J. Geophys. Res.*, **109**, D16207
- 23 Liao, T., C.D. Camp, and Y.L. Yung, 2004: The seasonal cycle of N<sub>2</sub>O. *Geophys. Res. Lett.*, **31**, L17108.
- 24 Liousse, C., et al., 1996: A global three-dimensional model study of carbonaceous aerosols. *J. Geophys.*  
25 *Res.*, **101**(D14), 19,411-19,432.
- 26 Liu, H.Q., R.T. Pinker, and B.N. Holben, 2005: A global view of aerosols from merged transport models,  
27 satellite, and ground observations. *J. Geophys. Res.*, **110**(D10), doi:10.1029/2004JD004695.
- 28 Liu, X.H., and J.E. Penner, 2002: Effect of Mount Pinatubo H<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O aerosol on ice nucleation in the  
29 upper troposphere using a global chemistry and transport model. *J. Geophys. Res.*, **107**(D12),  
30 doi:10.1029/2001JD000455.
- 31 Liu, Y., and P.H. Daum, 2002: Indirect warming effect from dispersion forcing. *Nature*, **419**, 580-581.
- 32 Lockwood, M., and R. Stamper, 1999: Long-term drift of the coronal source magnetic flux and the total solar  
33 irradiance. *Geophys. Res. Lett.*, **26**(16), 2461-2464.
- 34 Loeb, N.G., and S. Kato, 2002: Top-of-atmosphere direct radiative effect of aerosols over the tropical oceans  
35 from the Clouds and the Earth's Radiant Energy System (CERES) satellite instrument. *J. Climate*,  
36 **15**, 1474-1484.
- 37 Loeb, N.G., and N. Manalo-Smith, 2005: Top-of-atmosphere direct radiative effect of aerosols over global  
38 oceans from merged CERES and MODIS observations. *J. Climate*, **18**, 3506-3526.
- 39 Lohar, D., and B. Pal, 1995: The effect of irrigation on premonsoon season precipitation over south west  
40 Bengal, India. *J. Climate*, **8**, 2567-2570.
- 41 Lohmann, U., and G. Lesins, 2002: Stronger constraints on the anthropogenic indirect aerosol effect.  
42 *Science*, **298**, 1012-1016.
- 43 Lohmann, U., and J. Feichter, 2005: Global indirect aerosol effects: A review. *Atmos. Chem. Phys.*, **5**, 715-  
44 737.
- 45 Lohmann, U. and C. Leck, 2005: Importance of submicron surface active organic aerosols for pristine Arctic  
46 clouds. *Tellus*, **57B**, 261-268.
- 47 Lohmann, U., J. Feichter, J.E. Penner, and W.R. Leaitch, 2000: Indirect effect of sulfate and carbonaceous  
48 aerosols: a mechanistic treatment. *J. Geophys. Res.*, **105**(D10), 12,193-12,206.
- 49 Lohmann, U., et al., 2001: Vertical distributions of sulfur species simulated by large scale atmospheric  
50 models in COSAM: comparison with observations. *Tellus*, **53B**, 646-672.
- 51 Lohmann, U., B. Kärcher, and J. Hendrichs, 2004: Sensitivity studies of cirrus clouds formed by  
52 heterogeneous freezing in ECHAM GCM. *J. Geophys. Res.*, **109**, D16204, doi:  
53 10.1029/2003JD004443.
- 54 Loveland, T.R., et al., 2000: Development of a global land cover characteristics database and IGBP  
55 DISCover from 1 km AVHRR R data. *Int. J. Remote Sens.*, **21**, 1303-1330.
- 56 Lowe, D.C., and W. Allan, 2002: A simple procedure for evaluating global cosmogenic <sup>14</sup>C production in the  
57 atmosphere using neutron monitor data. *Radiocarbon*, **44**, 149-157.

- 1 Lowe, D.C., M.R. Manning, G.W. Brailsford, and A.M. Bromley, 1997: The 1991-1992 atmospheric  
2 methane anomaly: Southern hemisphere <sup>13</sup>C decrease and growth rate fluctuations. *Geophys. Res.  
3 Lett.*, **24**(8), 857-860.
- 4 Lowe, D.C., et al., 2004: Seasonal cycles of mixing ratio and 13C in atmospheric methane at Suva, Fiji. *J.  
5 Geophys. Res.*, **109**, D23308 doi:10.1029/2004JD005166.
- 6 Lu, M-H and J. Seinfeld, 2005: Study of the aerosol indirect effect by large-eddy simulation of marine  
7 stratocumulus. *J. Atmos. Sci.*, **62**, 3909-3932.
- 8 Luo C., N. M. Mahowald, J. del Corral, 2003: Sensitivity study of meteorological parameters on mineral  
9 aerosol mobilization, transport, and distribution, *J. Geophys. Res.*, **108** (D15), 4447,  
10 doi:10.1029/2003JD003483.
- 11 MacFarling Meure, C., et al., 2006: The Law Dome CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O ice core records extended to 2000  
12 years BP. *Geophys. Res. Lett.* **33**, L14810, doi:10.1029/2006GL026152.
- 13 Macke, A., M. Mishchenko, and B. Cairns, 1996: The influence of inclusions on light scattering by large ice  
14 particles. *J. Geophys. Res.*, **101**(D18), 23311-23316.
- 15 Magi, B.I., and P.V. Hobbs, 2003: Effects of humidity on aerosols in southern Africa during the biomass  
16 burning season. *J. Geophys. Res.*, **108**(D13), 8495, doi:10.1029/2002JD002144.
- 17 Magi, B.I., et al., 2005: Properties and chemical apportionment of aerosol optical depth at locations off the  
18 U.S. East Coast in July and August 2001. *J. Atmos. Sci.*, **62**(4), 919-933, doi: 10.1175/JAS3263.1.
- 19 Mahowald, N.M., G.C. Rivera, and C. Luo, 2004: Comment on Tegen et al. 2004, on the "Relative  
20 importance of climate and land use in determining present and future global soil dust emissions".  
21 *Geophys. Res. Lett.*, **31**, L24105, doi:10.1029/2004GL021272.
- 22 Mahowald, N.M., and C. Luo, 2003: A less dusty future? *Geophys. Res. Lett.*, **30**(17),  
23 doi:10.1029/2003GL017880.
- 24 Mak, J.E., M.R. Manning, and D.C. Lowe, 2000: Aircraft observations of δ<sup>13</sup>C of atmospheric methane over  
25 the Pacific in August 1991 and 1993: evidence of an enrichment in <sup>13</sup>CH<sub>4</sub> in the Southern  
26 Hemisphere. *J. Geophys. Res.* **105**(D1), 1329-1335.
- 27 Malm W.C., B.A. Schichtel, M.L. Pitchford, L.L. Ashbaugh, and R.A. Eldred, 2004: Spatial and monthly  
28 trends in speciated fine particle concentration in the United States. *J. Geophys. Res.*, **109**, D03306.
- 29 Manabe, S., and R.T. Wetherald, 1967: Thermal equilibrium of the atmosphere with a given distribution of  
30 relative humidity. *J. Atmos. Sci.*, **24**, 241-259.
- 31 Manne, A.S., and R.G. Richels, 2001: An alternative approach to establishing trade-offs among greenhouse  
32 gases. *Nature*, **410**, 675-676.
- 33 Manning, A.C., and R.F. Keeling, 2006: Global oceanic and land biotic carbon sinks from the Scripps  
34 atmospheric oxygen flask sampling network. *Tellus*, **58B**, 95-116.
- 35 Manning, A.J., D.B. Ryall, R.G. Derwent, P.G. Simmonds, and S. O Doherty, 2003: Estimating European  
36 emissions of ozone-depleting and greenhouse gases using observations and a modelling back-  
37 attribution technique. *J. Geophys. Res.*, **108** (D14), 4405, doi:10.1029/2002JD002312.
- 38 Manning, M.R., A. Gomez, and G.W. Brailsford, 1997: Annex B11: The New Zealand CO<sub>2</sub> measurement  
39 programme. In: *Report of the Ninth WMO Meeting of Experts on Carbon Dioxide Concentration and  
40 Related Tracer Measurement Techniques* (WMO Global Atmosphere Watch No. 132 ; WMO TD  
41 No. 952). CSIRO, Melbourne, pp. 120-123.
- 42 Manning, M.R., D.C. Lowe, R.C. Moss, G.E. Bodeker, and W. Allan, 2005: Short term variations in the  
43 oxidizing power of the atmosphere. *Nature*, **436**, 1001-1004.
- 44 Mannstein, H., and U. Schumann, 2005: Aircraft induced contrail cirrus over Europe. *Meteorol. Z.*, **14**, 549-  
45 544.
- 46 Maria, S., F. Russell, L.M. Turpin, and R.J. Porcja, 2002: FTIR measurements of functional groups and  
47 organic mass in aerosol samples over the Caribbean. *Atmos. Environ.*, **36**, 5185-5196.
- 48 Marland, G., T.A. Boden, and R.J. Andres, 2006: Global, regional, and national CO<sub>2</sub> emissions. In *Trends: A  
49 Compendium of Data on Global Change. Carbon Dioxide Information Analysis Center, Oak Ridge  
50 National Laboratory, U.S. Department of Energy, Oak Ridge, Tenn., U.S.A.*  
51 [http://cdiac.esd.ornl.gov/trends/emis/tre\\_glob.htm](http://cdiac.esd.ornl.gov/trends/emis/tre_glob.htm)
- 52 Marquart, S., and B. Mayer, 2002: Towards a reliable GCM estimation of contrail radiative forcing.  
53 *Geophys. Res. Lett.*, **29**(8), doi: 10.1029/2001GL014075.
- 54 Marquart, S., M. Ponater, F. Mager, and R. Sausen, 2003: Future development of contrail cover, optical  
55 depth, and radiative forcing: Impacts of increasing air traffic and climate change. *J. Climate*, **16**,  
56 2890-2904.

- 1 Marsh, N.D. and H. Svensmark, 2000a: Low cloud properties influenced by cosmic rays. *Phys. Rev. Lett.*,  
2 **85**, 5004–5007.
- 3 Marsh, N.D. and H. Svensmark, 2000b: Cosmic rays, clouds, and climate. *Space Science Rev.*, **94**, 215–230.
- 4 Marshak A., S. Platnick, T. Várnai, G. Wen, and R.F. Cahalan, 2006: Impact of three-dimensional radiative  
5 effects on satellite retrievals of cloud droplet sizes. *J. Geophys. Res.*, **111**, D09207,  
6 doi:10.1029/2005JD006686.
- 7 Martin, G. M., D.W. Johnson, and A.Spice, 1994: The measurement and parameterization of effective radius  
8 of droplets in warm stratiform clouds. *J. Atmos. Sci.*, **51**, 1823–1842.
- 9 Martin, S.T., et al., 2004: Effects of the physical state of tropospheric ammonium-sulfate-nitrate particles on  
10 global aerosol direct radiative forcing. *Atmos. Chem. Phys.*, **4**, 183-214.
- 11 Martonchik, J.V., D.J. Diner, R. Kahn, B. Gaitley, and B. Holben, 2004: Comparison of MISR and  
12 AERONET aerosol optical depths over desert sites. *Geophys. Res. Lett.*, **31**, L16102,  
13 doi:10.1029/2004GL019807.
- 14 Matthews, E., 1983: Global vegetation and land-use: new high-resolution data-bases for climate studies. *J.*  
15 *Clim. Appl. Meteorol.*, **22**, 474-487.
- 16 Matthews, H.D., A.J. Weaver, M. Eby, and K.J. Meissner, 2003: Radiative forcing of climate by historical  
17 land cover change. *Geophys. Res. Lett.*, **30**(2), 271-274.
- 18 Matthews, H.D., A.J. Weaver, K.J. Meissner, N.P. Gillett, and M. Eby, 2004: Natural and anthropogenic  
19 climate change: Incorporating historical land cover change, vegetation dynamics and the global  
20 carbon cycle. *Clim. Dyn.*, **22**, 461-479.
- 21 Matthias, I., A. Ansmann, D. Müller, U. Wandinger, and D. Althausen, 2004: Multiyear aerosol observations  
22 with dual wavelength Raman lidar in the framework of EARLINET. *J. Geophys. Res.*, **109**, D13203,  
23 doi:10.1029/2004JD004600.
- 24 McCormack, J.P., 2003: The influence of the 11-year solar cycle on the quasi-biennial oscillation. *Geophys.*  
25 *Res. Lett.*, **30** (22), 2162, doi:10.1029/2003GL018314.
- 26 McCormick, M. P., 1987: SAGE II: An overview. *Adv. Space Res.*, **7**, 219-226.
- 27 McCulloch, A., and P.M. Midgley, 2001: The history of methyl chloroform emissions: 1951-2000. *Atmos.*  
28 *Env.*, **35**, 5311-5319.
- 29 McFiggans, G., et al., 2006: The effect of aerosol composition and properties on warm cloud droplet  
30 activation. *Atmos. Chem. Phys.*, **6**, 2593–2649.
- 31 Meehl, G.A., et al., 2004: Combinations of natural and anthropogenic forcings in twentieth-century climate.  
32 *J. Climate*, **17**, 3721-3727.
- 33 Meerkötter, R., et al., 1999: Radiative forcing by contrails. *Ann. Geophysicae*, **17**, 1080-1094.
- 34 Melillo, J.M., et al. 2001: Nitrous oxide emissions from forests and pastures of various ages in the Brazilian  
35 Amazon. *J. Geophys. Res.*, **106**(D24), 34,179-34,188.
- 36 Menon, S., and A. Del Genio, 2006: Evaluating the impacts of carbonaceous aerosols on clouds and climate.  
37 In: Human-Induced Climate Change: an Interdisciplinary Assessment [M. Schlesinger et al. (eds.).  
38 Cambridge Univ. Press, Cambridge, UK (in press)
- 39 Menon, S., J. Hansen, L. Nazarenko, and Y. Luo, 2002a: Climate effects of black carbon aerosols in China  
40 and India. *Science*, **297**, 2250-2253.
- 41 Menon, S., A.D. Del Genio, D. Koch, and G. Tselioudis, 2002b: GCM simulations of the aerosol indirect  
42 effect: sensitivity to cloud parameterization and aerosol burden. *J. Atmos. Sci.*, **59**, 692-713.
- 43 Menon, S., et al., 2003: Evaluating aerosol/cloud/radiation process parameterizations with single-column  
44 models and Second Aerosol Characterization Experiment (ACE-2) cloudy column observations. *J.*  
45 *Geophys. Res.*, **108**(D24), 4762, doi:10.1029/2003JD003902.
- 46 Meyer, R., H. Mannstein, R. Meerkötter, U. Schumann, and P. Wendling, 2002: Regional radiative forcing  
47 by line-shaped contrails derived from satellite data. *J. Geophys. Res.*, **107**(D10), 4104, doi:  
48 10.1029/2001JD000426.
- 49 Mickley, L.J., D.J. Jacob, and D. Rind, 2001: Uncertainty in preindustrial abundance of tropospheric ozone:  
50 implications for radiative forcing calculations. *J. Geophys. Res.*, **106**(D4), 3389-3399  
51 doi:10.1029/2000JD900594.
- 52 Mickley, L.J., D.J. Jacob, B.D. Field, and D. Rind, 2004: Climate response to the increase in tropospheric  
53 ozone since preindustrial times: a comparison between ozone and equivalent CO2 forcings. *J.*  
54 *Geophys. Res.*, **109**, D05106, doi:10.1029/2003JD003653.
- 55 Mikami, M., et al., 2006: Aeolian dust experiment on climate impact: an overview of Japan-China Joint  
56 Project ADEC, Global and Planetary Change, **52**, 142-172. doi:10.1016/j.gloplacha.2006.03.001.

- 1 Miller, B.R., J. Huang, R.F. Weiss, R.G. Prinn, and P.J. Fraser, 1998: Atmospheric trend and lifetime of  
2 chlorodifluoromethane (HCFC-22) and the global tropospheric OH concentration. *J. Geophys. Res.*,  
3 **103**(D11), 13237-13248, doi: 10.1029/98JD00771.
- 4 Miller, R.L., G.A. Schmidt, and D.T. Shindell, 2006: Forced Variations of Annular Modes in the 20th  
5 Century IPCC AR4 Simulations. *J. Geophys. Res.*, doi 2005JD006323.
- 6 Millet, D.B., and A.H. Goldstein, 2004: Evidence of continuing methylchloroform emissions from the  
7 United States. *Geophys. Res. Lett.*, **31**, L17101-17106.
- 8 Ming, Y., and L.M. Russell, 2002: Thermodynamic equilibrium of organic-electrolyte mixtures in aerosol  
9 particles. *Am. Inst. Chem. Eng. J.*, **48**, 1331-1348.
- 10 Ming, Y., V. Ramaswamy, P.A. Ginoux, and L.H. Horowitz, 2005a: Direct radiative forcing of  
11 anthropogenic organic aerosol. *J. Geophys. Res.*, **110**, D20208, doi:10.1029/2004JD005573.
- 12 Ming, Y., V. Ramaswamy, P.A. Ginoux, L.W. Horowitz, and L.M. Russell, 2005b: Geophysical Fluid  
13 Dynamics Laboratory general circulation model investigation of the indirect radiative effects of  
14 anthropogenic sulfate aerosol. *J. Geophys. Res.*, **110**, D22206, doi:10.1029/2005JD006161.
- 15 Ming, Y., V. Ramaswamy, Leo J. Donner, and Vaughan T. J. Phillips, 2006a: A new parameterization of  
16 cloud droplet activation applicable to general circulation models. *J. Atmos. Sci.*, **63**(4), 1348-1356.
- 17 Ming, Y., V. Ramaswamy, L.J. Donner, V.T.J. Phillips, and S.A. Klein, 2006b: Modelling the interactions  
18 between aerosols and liquid water clouds with a self-consistent cloud scheme in a general circulation  
19 model. *J. Atmos. Sci.* (in press).
- 20 Minnis, P., 2005: Reply. *J. Climate*, **18**, 2783-2784.
- 21 Minnis, P., J.K. Ayers, R. Palikonda, and D. Phan, 2004: Contrails, cirrus trends, and climate. *J. Climate*, **17**,  
22 1671-1685.
- 23 Minnis, P., et al., 1998: Transformation of contrails into cirrus during SUCCESS. *Geophys. Res. Lett.*, **25**,  
24 1157-1160.
- 25 Minnis, P., et al., 1993: Radiative climate forcing by the Mt. Pinatubo eruption. *Science*, **259**, 1411-1415.
- 26 Mircea, M., et al., 2005: Importance of the organic aerosol fraction for modeling aerosol hygroscopic growth  
27 and activation: a case study in the Amazon Basin. *Atmos. Chem. Phys.*, **5**, 3111-3126.
- 28 Mishchenko, M.I., I.V. Geogdzhayev, B. Cairns, W.B. Rossow, and A.A. Lacis, 1999: Aerosol retrievals  
29 over the ocean by use of channels 1 and 2 AVHRR data: sensitivity analysis and preliminary results.  
30 *Appl. Opt.*, **38**, 7325-7341.
- 31 Möhler, O., et al., 2005: Effect of sulfuric acid coating on heterogenous ice nucleation by soot aerosol  
32 particles. *J. Geophys. Res.*, **110**, D11210, doi:10.1029/2004JD005169.
- 33 Montzka, S.A., et al., 1999: Present and future trends in the atmospheric burden of ozone-depleting halogens.  
34 *Nature*, **398**, 690-694.
- 35 Montzka, S.A., et al., 2003: Controlled substances and other source gases. In: *Scientific assessment of ozone*  
36 *depletion : 2002 : pursuant to Article 6 of the Montreal Protocol on Substances that Deplete the*  
37 *Ozone Layer / World Meteorological Organization*. WMO, Geneva, pp. 1.1-1.83.
- 38 Moore, N., and S. Rojstaczer, 2001: Irrigation-induced rainfall and the Great Plains. *J. Appl. Meteorol.*, **40**,  
39 1297-1309.
- 40 Morgan, C.G., et al., 2004: Isotopic fractionation of nitrous oxide in the stratosphere: Comparison between  
41 model and observations. *J. Geophys. Res.*, **109**, D04305, doi:10.1029/2003JD003402.
- 42 Morimoto S., S. Aoki, T. Nakazawa, T. Yamanouchi, 2006: Temporal variations of the carbon isotopic ratio  
43 of atmospheric methane observed at Ny Ålesund, Svalbard from 1996 to 2004. *Geophys. Res. Lett.*,  
44 **33**,L01807, doi:10.1029/2005GL024648
- 45 Mosley-Thompson, E., T.A. Mashiotta, and L.G. Thompson, 2003: High resolution ice core records of Late  
46 Holocene volcanism: Current and future contributions from the Greenland PARCA cores, In:  
47 *Volcanism and the Earth's Atmosphere* [Robock, A. and C. Oppenheimer (eds.)], Geophysical  
48 Monograph 139, American Geophysical Union, pp. 153-164
- 49 Mouillot, F., A. Narasimha, Y. Balkanski, J.F. Lamarque, and C.B. Field, 2006: Global carbon emissions  
50 from biomass burning in the 20th century. *Geophys. Res. Lett.*, **33**, L01801.
- 51 Moulin, C., H.R. Gordon, V.F. Banzon, and R.H. Evans, 2001: Assessment of Saharan dust absorption in the  
52 visible from SeaWiFS imagery. *J. Geophys. Res.*, **106**(D16), 18239-18249.
- 53 Moulin C., I. Chiapello, 2004: Evidence of the control of summer atmospheric transport of African dust over  
54 the Atlantic by Sahel sources from TOMS satellites (1979-2000). *Geophys. Res. Lett.* **31**, L02107
- 55 Murayama, T., et al., 2001: Ground-based network observation of Asian dust events of April 1998 in east  
56 Asia. *J. Geophys. Res.*, **106**, 18345-18360.
- 57 Murphy, D.M., 2005: Something in the air. *Science*, **307**, 1888-1890.

- 1 Muscheler, R., et al., 2006: Solar activity during the last 1000 yr inferred from radionuclide records. *Quat.*  
2 *Sci. Rev.* doi:10.1016/j.quascirev.2006.07.012
- 3 Myhre, G., and F. Stordal, 2001a: Global sensitivity experiments of the radiative forcing due to mineral  
4 aerosols. *J. Geophys. Res.*, **106**, 18,193-18,204.
- 5 Myhre, G., and F. Stordal, 2001b: On the tradeoff of the solar and thermal infrared radiative impact of  
6 contrails. *Geophys. Res. Lett.*, **28**, 3119-3122.
- 7 Myhre, G., and A. Myhre, 2003: Uncertainties in radiative forcing due to surface albedo changes caused by  
8 land-use changes. *J. Climate*, **16**, 1511-1524.
- 9 Myhre, G., A. Myhre, and F. Stordal, 2001: Historical evolution of radiative forcing of climate. *Atmos. Env.*,  
10 **35**, 2361-2373.
- 11 Myhre, G., et al., 2003: Modelling the solar radiative impact of aerosols from biomass burning during the  
12 Southern African Regional Science Initiative (SAFARI 2000) experiment. *J. Geophys. Res.*, **108**,  
13 SAF 37, doi:10.1029/2002JD002313.
- 14 Myhre, G., et al., 2004a: Intercomparison of satellite retrieved aerosol optical depth over ocean, *J. Atmos.*  
15 *Sci.*, **61**, 499–513.
- 16 Myhre, G., F. Stordal, T.F. Berglen, J.K. Sundet, and I.S.A. Isaksen, 2004b: Uncertainties in the radiative  
17 forcing due to sulfate aerosols. *J. Atmos. Sci.*, **61**, 485-498.
- 18 Myhre, G., et al., 2005a: Intercomparison of satellite retrieved aerosol optical depth over ocean during the  
19 period September 1997 to December 2000. *Atmos. Chem. Phys.*, **5**, 1697-1719.
- 20 Myhre, G., M.M. Kvalevåg, and C.B. Schaaf, 2005b: Radiative forcing due to anthropogenic vegetation  
21 change based on MODIS surface albedo data. *Geophys. Res. Lett.*, **32**, L21410.
- 22 Naja, M., and H. Akimoto, 2004: Contribution of regional pollution and long-range transport to the Asia-  
23 Pacific region: Analysis of long-term ozonesonde data over Japan. *J. Geophys. Res.*, **109**, 1306,  
24 doi:10.1029/2004JD004687.
- 25 Naja, M., H. Akimoto, and J. Staehelin, 2003: Ozone in background and photochemically aged air over  
26 central Europe: analysis of long-term ozonesonde data from Hohenpeissenberg and Payerne. *J.*  
27 *Geophys. Res.*, **108**, 4063, doi:10.1029/2002JD002477.
- 28 Nakajima, T., T. Hayasaka, A. Higurashi, G. Hashida, N. Moharram-Nejad, Y. Najafi, and H. Valavi, 1996:  
29 Aerosol Optical Properties in the Iranian Region Obtained by Ground-Based Solar Radiation  
30 Measurements in the Summer of 1991. *J. Appl. Meteorol.*, **35**, 1265-1278.
- 31 Nakajima, T. and A. Higurashi, 1998: A use of two-channel radiances for an aerosol characterization from  
32 space. *Geophys. Res. Lett.*, **25**, 3815-3818.
- 33 Nakajima, T., A. Higurashi, K. Kawamoto and J. Penner, 2001: A possible correlation between satellite-  
34 derived cloud and aerosol microphysical parameters. *Geophys. Res. Lett.*, **28**, 1171-1174.
- 35 Nakazawa, T., S. Moromoto, S. Aoki, and M. Tanaka, 1997: Temporal and spatial variations of the carbon  
36 isotopic ratio of atmospheric carbon dioxide in the Western Pacific region. *J. Geophys. Res.*, **102**,  
37 1271-1285.
- 38 Nakicenovic, N., A. Grübler, and A. McDonald (eds). (1998). *Global energy: perspectives*. Cambridge  
39 University Press, New York. 299 p.
- 40 Nenes, A., and J.H. Seinfeld, 2003: Parameterization of cloud droplet formation in global climate models. *J.*  
41 *Geophys. Res.*, **108**, doi: 10.1029/2002JD002911.
- 42 Nenes, A., et al., 2002: Can chemical effects on cloud droplet number rival the first indirect effect? *Geophys.*  
43 *Res. Lett.*, **29**, 1848, doi:10.1029/2002GL015295.
- 44 Nevison, C.D., S. Solomon, R.S. Gao, 1999: Buffering interactions in the modeled response of stratospheric  
45 O<sub>3</sub> to increased NO<sub>x</sub> and HO<sub>x</sub>. *J. Geophys. Res.*, **104**(D3), 3741-3754, 10.1029/1998JD100018.
- 46 Nevison, C.D., D.E. Kinnison, and R.F. Weiss, 2004a: Stratospheric influences on the tropospheric seasonal  
47 cycles of nitrous oxide and chlorofluorocarbons. *Geophys. Res. Lett.*, **31**, L20103,  
48 doi:10.1029/2004GL020398.
- 49 Nevison, C., T. Lueker, and R.F. Weiss, 2004b: Quantifying the nitrous oxide source from coastal upwelling,  
50 *Global Biogeochem. Cycles*, **18**, GB1018, doi:10.1029/2003GB002110.
- 51 Nevison, C.D., et al., 2005: Southern Ocean ventilation inferred from seasonal cycles of atmospheric N<sub>2</sub>O  
52 and O<sub>2</sub>/N<sub>2</sub> at Cape Grim, Tasmania. *Tellus*, **57B**, 218-229.
- 53 Newchurch, M.J., et al., 2003: Evidence for slowdown in stratospheric ozone loss: first stage of ozone  
54 recovery. *J. Geophys. Res.*, **108**(D16), 4507, doi:10.1029/2003JD003471.
- 55 Nordhaus, W.D., 1997: Discounting in economics and climate change: an editorial comment. *Clim. Change*,  
56 **37**, 315-328.

- 1 Notholt, J., et al., 2005: Influence of tropospheric SO<sub>2</sub> emissions on particle formation and the stratospheric  
2 humidity. *Geophys. Res. Lett.*, **32**, L07810, doi:10.1029/2004GL022159.
- 3 Nozawa, T, T. Nagashima, H. Shiogama, and S.A. Crooks, 2005: Detecting natural influence on surface air  
4 temperature change in the early twentieth century. *Geophys. Res. Lett.*, **32**, L20719,  
5 doi:10.1029/2005GL023540
- 6 Novakov, T., D.A. Hegg, and P.V. Hobbs, 1997: Airborne measurements of carbonaceous aerosols on the  
7 East coast of the United States. *J. Geophys. Res.*, **102**(D25), 30023-20030.
- 8 Novakov, T., et al., 2003: Large historical changes of fossil-fuel black carbon aerosols. *Geophys. Res. Lett.*,  
9 **30**(6), 1324, doi:10.1029/2002GL016345.
- 10 O'Doherty, S., et al., 2004: Rapid growth of hydrofluorocarbon 134a and hydrochlorofluorocarbons 141b,  
11 142b, and 22 from Advanced Global Atmospheric Gases Experiment (AGAGE) observations at  
12 Cape Grim, Tasmania, and Mace Head, Ireland. *J. Geophys. Res.*, **109**, D06310,  
13 doi:10.1029/2003JD004277.
- 14 O'Neill, B.C. 2000: The jury is still out on global warming potentials. *Clim. Change*, **44**, 427-443.
- 15 O'Neill, B. 2003: Economics, natural science, and the costs of global warming potentials. *Clim. Change*, **58**,  
16 251-260.
- 17 Oinas, V., A.A. Lacis, D. Rind, D.T. Shindell, and J.E. Hansen, 2001: Radiative cooling by stratospheric  
18 water vapor: big differences in GCM results. *Geophys. Res. Lett.*, **28**, 2791-2794.
- 19 Olivier, J.G.J., and J.J.M. Berdowski, 2001: Global emissions sources and sinks. In: *The Climate System*.  
20 [Berdowski, J. R. Guicherit, and B.J. Heij (eds.)]. A.A. Balkema/Swets & Zeitlinger, Lisse, The  
21 Netherlands, pp. 33-78, updated at <http://www.mnp.nl/edgar/>.
- 22 Oltmans, S.J., et al., 2006: Long-term changes in tropospheric ozone. *Atmos. Environ.*, **40**, 3156-317.
- 23 Oram, D.E., 1999: *Trends of long-lived anthropogenic halocarbons in the Southern Hemisphere and model*  
24 *calculations of global emissions*. Ph.D. Thesis, University of East Anglia, Norwich, U.K. p.249.
- 25 Oram, D.E., W.T. Sturges, S.A. Penkett, A. McCulloch, and P.J. Fraser, 1998: Growth of fluoroform (CHF<sub>3</sub>,  
26 HFC-23) in the background atmosphere. *Geophys. Res. Lett.*, **25**, 35-38, doi: 10.1029/97GL03483.
- 27 Osborne, S.R., J.M. Haywood, P.N. Francis, and O. Dubovik, 2004: Short-wave radiative effects of biomass  
28 burning aerosol during SAFARI2000. *Q.J.R. Meteorol. Soc.*, **130**, 1423-1448.
- 29 Palikonda, R., P. Minnis, D.P. Duda, and H. Mannstein, 2005: Contrail coverage derived from 2001 AVHRR  
30 data over the continental United States of America and surrounding area. *Meteorol. Z.*, **14**, 525-536.
- 31 Palmer, P.I., et al., 2003: Eastern Asian emissions of anthropogenic halocarbons deduced from aircraft  
32 concentration data. *J. Geophys. Res.*, **108**(D24), 4753, doi:10.1029/2003JD003591.
- 33 Palmer, A.S., V.I. Morgan, A.J. Curran, T.D. Van Ommen, and P.A. Mayewski, 2002: Antarctic volcanic  
34 flux ratios from Law Dome ice cores. *Annals of Glaciology*, **35**, 329-332.
- 35 Pan, W., M.A. Tatang, G.J. McRae, and R.G. Prinn, 1997: Uncertainty analysis of direct radiative forcing by  
36 anthropogenic sulfate aerosols. *J. Geophys. Res.*, **102**(D18), 21,915-21,924.
- 37 Parrish, D.D., et al., 2004: Fraction and composition of NO<sub>y</sub> transported in air masses lofted from the North  
38 American continental boundary layer. *J. Geophys. Res.*, **109**, D09302, doi:10.1029/2003JD004226.
- 39 Pavelin, E.G., C.E. Johnson, S. Rughooputh, and R. Toumi, 1999: Evaluation of pre-industrial surface ozone  
40 measurements made using Schönbein's method. *Atmos. Environ.*, **33**, 919-929
- 41 Peng, Y., and U. Lohmann, 2003: Sensitivity study of the spectral dispersion of the cloud droplet size  
42 distribution on the indirect aerosol effect. *Geophys. Res. Lett.*, **30**(10), 1507,  
43 doi:10.1029/2003GL017192.
- 44 Penner, J.E., et al., 2001: Chapter 5: Aerosols, their direct and indirect effects. In: *Climate Change 2001: The*  
45 *Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the*  
46 *Intergovernmental Panel on Climate Change* [Houghton, J.T., et al. (eds.)]. Cambridge University  
47 Press, Cambridge, U.K, pp. 289-348.
- 48 Penner, J. E., et al., 2002: A comparison of model- and satellite-derived aerosol optical depth and  
49 reflectivity. *J. Atmos. Sci.*, **59**, 441-460.
- 50 Penner, J.E., S.Y. Zhang, and C.C. Chuang, 2003: Soot and smoke aerosol may not warm climate. *J.*  
51 *Geophys. Res.*, **108**(D21), 4657, doi:10.1029/2003JD003409.
- 52 Penner, J.E., X. Dong, and Y. Chen, 2004: Observational evidence of a change in radiative forcing due to the  
53 indirect aerosol effect. *Nature*, **427**, 231-234.
- 54 Penner, J.E., et al., 2006a: Model intercomparison of indirect aerosol effects. *Atmos. Chem. Phys. Discuss.*,  
55 **6**, 1579-1617.

- 1 Penner, J.E., M. Wang, A. Kumar, L. Rotstayn, and B. Santer, 2006b: Effect of black carbon on mid-  
2 troposphere and surface temperature trends. In: Human-Induced Climate Change: an  
3 Interdisciplinary Assessment, [M. Schlesinger, et al., (eds.)]. Cambridge University Press, (in press)
- 4 Perlwitz, J., and H.-F. Graf, 2001: Troposphere-stratosphere dynamic coupling under strong and weak polar  
5 vortex conditions. *Geophys. Res. Lett.*, **28**, 271-274.
- 6 Perlwitz, J., and N. Harnik, 2003: Observational evidence of a stratospheric influence on the troposphere by  
7 planetary wave reflection. *J. Climate*, **16**, 3011-3026.
- 8 Parrish, D. D., et al., 2004: Changes in the photochemical environment of the temperate North Pacific  
9 troposphere in response to increased Asian emissions. *J. Geophys. Res.*, **109**, D23S18,  
10 doi:10.1029/2004JD004978.
- 11 Pham, M., O. Boucher, and D. Hauglustaine, 2005: Changes in atmospheric sulfur burdens and  
12 concentrations and resulting radiative forcings under IPCC SRES emission scenarios for 1990-2100.  
13 *J. Geophys. Res.*, **110**, D06112, doi:10.1029/2004JD005125.
- 14 Philipona, R., B. Durr, C. Marty, A. Ohmura, and M. Wild, 2004: Radiative forcing - measured at Earth's  
15 surface - corroborate the increasing greenhouse effect. *Geophys. Res. Lett.*, **31**, L03202,  
16 doi:10.1029/2003GL018765.
- 17 Pielke Sr., R.A., et al., 2002: The influence of land-use change and landscape dynamics on the climate  
18 system - relevance to climate change policy beyond the radiative effect of greenhouse gases. *Philos.*  
19 *Trans. R. Soc. London Ser. A*, **360**, 1705-1719.
- 20 Pitari, G., E. Mancini, V. Rizi, and D.T. Shindell, 2002: Impact of future climate and emissions changes on  
21 stratospheric aerosols and ozone. *J. Atmos. Sci.*, **59**, 414-440.
- 22 Platt, U., W. Allan, and D.C. Lowe, 2004: Hemispheric average Cl atom concentration from <sup>13</sup>C/<sup>12</sup>C ratios in  
23 atmospheric methane. *Atmos. Chem. Phys.*, **4**, 2393-2399.
- 24 Ponater, M., S. Marquart, and R. Sausen, 2002: Contrails in a comprehensive global climate model:  
25 Parameterization and radiative forcing results. *J. Geophys. Res.*, **107**(D13), doi:  
26 10.1029/2001JD000429.
- 27 Ponater, M., S. Marquart, R. Sausen, and U. Schumann, 2005: On contrail climate sensitivity. *Geophys. Res.*  
28 *Lett.*, **32**, L10706, doi:10.1029/2005GL022580.
- 29 Posfai, M., R. Simonics, J. Li, P.V. Hobbs, and P.R. Buseck, 2003: Individual aerosol particles from biomass  
30 burning in southern Africa: 1. Compositions and size distributions of carbonaceous particles. *J.*  
31 *Geophys. Res.*, **108**(D13), 8483, doi:10.1029/2002JD002291.
- 32 Prata, A., W. Rose, S. Self, and D. O'Brien, 2003: Global, long-term sulphur dioxide measurements from  
33 TOVS data: a new tool for studying explosive volcanism and climate. In: *Volcanism and the Earth's*  
34 *Atmosphere* [Robock, A., and C. Oppenheimer (eds.)], Geophysical Monograph 139, American  
35 Geophysical Union, Washington, D.C., pp. 75-92.
- 36 Prather, M.J., 2003: Atmospheric science: an environmental experiment with H<sub>2</sub>? *Science*, **302**, 581-583.
- 37 Prather, M.J., et al., 2001: Atmospheric chemistry and greenhouse gases. In: *Climate Change 2001: The*  
38 *Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the*  
39 *Intergovernmental Panel on Climate Change* [Houghton, J.T., et al. (eds.)]. Cambridge University  
40 Press, Cambridge, U.K. pp. 239-287.
- 41 Preminger, D. G. and S. R. Walton, 2005: A new model of total solar irradiance based on sunspot areas,  
42 *Geophysical Research Letters*, **32**, L14109, doi:10.1029/2005GL022839
- 43 Prentice, I.C. et al., 2001: The carbon cycle and atmospheric carbon dioxide. In: *Climate Change 2001: The*  
44 *Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the*  
45 *Intergovernmental Panel on Climate Change* [Houghton, J.T., et al. (eds.)]. Cambridge University  
46 Press, Cambridge, U.K. pp. 184-238.
- 47 Prinn, R.G., et al., 1990: Atmospheric emissions and trends of nitrous oxide deduced from ten years of ALE-  
48 GAGE data. *J. Geophys. Res.*, **95**, 18369-18385.
- 49 Prinn, R.G., et al., 2000: A history of chemically and radiatively important gases in air deduced from  
50 ALE/GAGE/AGAGE. *J. Geophys. Res.*, **105**(D14), 17751-17792.
- 51 Prinn, R.G., et al., 2001: Evidence for substantial variations of atmospheric hydroxyl radicals in the past two  
52 decades. *Science*, **292**, 1882-1888.
- 53 Prinn, R.G., 2004: Non-CO<sub>2</sub> greenhouse gases, in *The Global Carbon Cycle* [C. Field and M. Raupach].  
54 Island Press, Washington, D.C., pp. 205-216, 2004.
- 55 Prinn, R.G., et al., 2005a: Evidence for variability of atmospheric hydroxyl radicals over the past quarter  
56 century. *Geophys. Res. Lett.*, **32**, L07809, doi:10.1029/2004GL022228.

- 1 Prinn, R.G., et al., 2005b: *The ALE/GAGE/AGAGE network: DB1001*. CDIAC World Data  
2 Center.<http://cdiac.esd.ornl.gov/ndps/alegage.html>.
- 3 Prospero, J.M., P. Ginoux, O. Torres, S.E. Nicholson, and T.E. Gill, 2002: Environmental characterization of  
4 global sources of atmospheric soil dust identified with the Nimbus 7 Total Ozone Mapping  
5 Spectrometer (TOMS) absorbing aerosol product. *Rev. Geophys.*, **40**(1),  
6 doi:10.1029/2000RG000095.
- 7 Putaud, J.P., et al., 2004: European aerosol phenomenology-2: chemical characteristics of particulate matter  
8 at kerbside, urban, rural and background sites in Europe. *Atmos. Environ.*, **38**, 2579-2595.
- 9 Pyle, J., et al., 2005: Ozone and climate: a review of interconnections. Compounds. In: *Special Report on*  
10 *Safeguarding the Ozone Layer and Global Climate System*. IPCC/TEAP, Cambridge UK, 83-132.
- 11 Quaas, J., O. Boucher, and F.-M. Breon, 2004: Aerosol indirect effects in POLDER satellite data and the  
12 Laboratoire de Météorologie Dynamique-Zoom (LMDZ) general circulation model. *J. Geophys.*  
13 *Res.*, **109**, D08205, doi:10.1029/2003JD004317.
- 14 Quaas J. and O. Boucher, 2005: Constraining the first aerosol indirect radiative forcing in the LMDZ GCM  
15 using POLDER and MODIS satellite data. *Geophys. Res. Lett.*, **32**, L17814,  
16 doi:10.1029/2005GL023850.
- 17 Quaas J., O. Boucher and U. Lohmann, 2005: Constraining the total aerosol indirect effect in the LMDZ and  
18 ECHAM4 GCMs using MODIS satellite data. *Atmos. Chem. Phys.*, **5**, 9669-9690.
- 19 Quay, P., et al., 2000: Atmospheric (CO)-C-14: A tracer of OH concentration and mixing rates. *J. Geophys.*  
20 *Res.*, **105**(D12), 15147-15166.
- 21 Quinn, P.K., and T.S. Bates, 2005: Regional aerosol properties: comparisons from ACE 1, ACE 2,  
22 Aerosols99, INDOEX, ACE Asia, TARFOX, and NEAQS. *J. Geophys. Res.*, **110**, D14202,  
23 doi:10.1029/2004JD004755.
- 24 Raes, F., T. Bates, F. McGovern, and M. Van Liedekerke, 2000: The 2nd Aerosol Characterization  
25 Experiment (ACE-2): general overview and main results. *Tellus*, **52B**, 111-125.
- 26 Ramachandran, S., V. Ramaswamy, G.L. Stenchikov, and A. Robock, 2000: Radioactive impact of the Mt.  
27 Pinatubo volcanic eruption: Lower stratospheric response. *J. Geophys. Res.*, **105**(D19), 24,409-  
28 24,429.
- 29 Ramanathan, V., 1981: The role of ocean-atmosphere interactions in the CO<sub>2</sub> climate problem. *J. Atmos. Sci.*,  
30 **38**, 918-930.
- 31 Ramanathan, V., P.J. Crutzen, J.T. Kiehl, and D. Rosenfeld, 2001a: Atmosphere: aerosols, climate, and the  
32 hydrological cycle. *Science*, **294**, 2119-2124.
- 33 Ramanathan, V., et al., 2001b: Indian Ocean experiment: An integrated analysis of the climate forcing and  
34 effects of the great Indo-Asian haze. *J. Geophys. Res.*, **106**(D22), 28371-28398.
- 35 Ramankutty, N., and J.A. Foley, 1999: Estimating historical changes in global land cover: croplands from  
36 1700 to 1992. *Global Biogeochem. Cycles*, **14**, 997-1027.
- 37 Ramaswamy, V., et al., 2001: Radiative forcing of climate change. In: *Climate Change 2001: The Scientific*  
38 *Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental*  
39 *Panel on Climate Change* [Houghton, J.T., et al. (eds.)]. Cambridge University Press, Cambridge,  
40 U.K, pp. 349-416.
- 41 Ramaswamy, V., et al., 2006a: Anthropogenic and natural influences in the evolution of lower stratospheric  
42 cooling. *Science*, **311**, 1138-1141.
- 43 Ramaswamy, V., S. Ramachandran, G. Stenchikov, and A. Robock, 2006: A model study of the effect of  
44 Pinatubo volcanic aerosols on the stratospheric temperatures. In: *Frontiers of Climate Modeling* [J.  
45 T. Kiehl, and V. Ramanathan, eds.] Cambridge University Press, Cambridge, U.K., pp. 152-178.
- 46 Randall, C.E., R.M. Bevilacqua, J.D. Lumpe, and K.W. Hoppel, 2001: Validation of POAM III aerosols:  
47 Comparison to SAGE II and HALOE. *J. Geophys. Res.*, **106**, 27,525-27,536.
- 48 Randles, C.A., L.M. Russell, and V. Ramaswamy, 2004: Hygroscopic and optical properties of organic sea  
49 salt aerosol and consequences for climate forcing. *Geophys. Res. Lett.*, **31**, L16108,  
50 doi:10.1029/2004GL020628.
- 51 Rasch, P.J., and J.E. Kristjánsson, 1998. A comparison of the CCM3 model climate using diagnosed and  
52 predicted condensate parameterizations. *J. Climate*, **11**, 1587- 1614.
- 53 Read, W.G., L. Froidevaux, and J.W. Waters, 1993: Microwave limb sounder measurements of stratospheric  
54 SO<sub>2</sub> from the Mt. Pinatubo volcano. *Geophys. Res. Lett.*, **20**(12), 1299-1302.
- 55 Reddy, M.S., and O. Boucher, 2004: A study of the global cycle of carbonaceous aerosols in the LMDZT  
56 general circulation model. *J. Geophys. Res.*, **109**, D14202, doi:10.1029/2003JD004048.

- 1 Reddy, M.S., O. Boucher, Y. Balanski, and M. Schulz, 2005a: Aerosol optical depths and direct radiative  
2 perturbations by species and source type. *Geophys. Res. Lett.*, **32**, L12803,  
3 doi:10.1029/2004GL021743.
- 4 Reddy, M.S., et al., 2005b: Estimates of global multicomponent aerosol optical depth and direct radiative  
5 perturbation in the Laboratoire de Météorologie Dynamique general circulation model. *J. Geophys.*  
6 *Res.*, **110**, D10S16, doi:10.1029/2004JD004757.
- 7 Reid, J.S., T.F. Eck, S.A. Christopher, P.V. Hobbs, and B. Holben, 1999: Use of the Angstrom exponent to  
8 estimate the variability of optical and physical properties of aging smoke particles in Brazil. *J.*  
9 *Geophys. Res.*, **104**(D22), 27,473-27,490.
- 10 Reid, J.S., et al., 2003: Analysis of measurements of Saharan dust by airborne and ground-based remote  
11 sensing methods during the Puerto Rico Dust Experiment (PRIDE). *J. Geophys. Res.*, **108**(D19),  
12 8586, doi:10.1029/2002JD002493.
- 13 Reimann, S., et al., 2005: Low methyl chloroform emissions inferred from long-term atmospheric  
14 measurements. *Nature*, **433**, 506-508, doi: 10.1038/nature03220.
- 15 Remer, L.A., and Y.J. Kaufman, 2006: Aerosol direct radiative effect at the top of the atmosphere over cloud  
16 free oceans derived from four years of MODIS data. *Atmos. Chem. Phys.*, **6**, 237-253.
- 17 Remer, L.A., et al., 2002: Validation of MODIS aerosol retrieval over ocean. *Geophys. Res. Lett.*, **29**(12),  
18 doi:10.1029/2001GL013204.
- 19 Remer, L. A., et al., 2005: The MODIS aerosol algorithm, products, and validation. *J. Atmos. Sci.*, **62**, 947-  
20 973.
- 21 Richards, J.F, 1990: Land transformation. In: *The Earth as Transformed by Human Action* [Turner II B.L., et  
22 al. (eds.)]. Cambridge University Press, New York, pp. 163-178.
- 23 Richter A., J.P., Burrows, H. Nüß, C. Granier, and U. Niemeier, 2005: Increase in tropospheric nitrogen  
24 dioxide over China observed from space. *Nature*, **437**, 129-132.
- 25 Rigozo, N.R., E. Echer, L.E.A. Vieira, and D.J.R. Nordemann, 2001: Reconstruction of Wolf sunspot  
26 numbers on the basis of spectral characteristics and estimates of associated radio flux and solar wind  
27 parameters for the last millennium. *Sol. Phys.*, **203**, 179-191.
- 28 Rind, D., J. Perlwitz, and P. Lonergan, 2005: AO/NAO response to climate change: 1. Respective influences  
29 of stratospheric and tropospheric climate changes. *J. Geophys. Res.*, **110**, D12107,  
30 doi:10.1029/2004JD005103.
- 31 Rissler, J., et al., 2004: Physical properties of the sub-micrometer aerosol over the Amazon rain forest during  
32 the wet-to-dry season transition – Comparison of modeled and measured CCN concentrations.  
33 *Atmos. Chem. Phys.*, **4**: 2119-2143.
- 34 Roberts, D.L., and A. Jones, 2004: Climate sensitivity to black carbon aerosol from fossil fuel combustion. *J.*  
35 *Geophys. Res.*, **109**, D16202, 1-12.
- 36 Robles-Gonzalez, C., J.P. Veefkind, and G. de Leeuw, 2000: Mean aerosol optical depth over Europe in  
37 August 1997 derived from ATSR-2 data. *Geophys. Res. Lett.* **27**(7), 955-959.
- 38 Robson J. I., L.K. Gohar, M.D. Hurley, K.P. Shine, and T.J. Wallington, 2006: Revised IR spectrum,  
39 radiative efficiency and global warming potential of nitrogen trifluoride. *Geophys. Res. Lett.*, **33**,  
40 L10817, doi:10.1029/2006GL026210
- 41 Rockmann, T., J. Grooss, and R. Muller, 2004: The impact of anthropogenic chlorine emissions,  
42 stratospheric ozone change and chemical feedbacks on stratospheric water. *Atmos. Chem. Phys.*, **4**,  
43 693-699.
- 44 Rosenfeld, D. and G. Feingold, 2003: Explanation of discrepancies among satellite observations of the  
45 aerosol indirect effects. *Geophys. Res. Lett.*, **30**(14), 1776, doi:10.1029/2003GL017684.
- 46 Rosenfeld, D., R. Lahav, K.A., and M. Pinsky, 2002: The role of sea spray in cleansing air pollution over the  
47 ocean via cloud processes. *Science*, **297**, 1667-1670.
- 48 Ross, K.E., et al., 2003: Spatial and seasonal variations in CCN distribution and the aerosol-CCN  
49 relationship over southern Africa. *J. Geophys. Res.*, **108**(D13), 8481, doi:10.1029/2002JD002384.
- 50 Rotstajn, L.D., 1997: A physically based scheme for the treatment of stratiform clouds and precipitation in  
51 large-scale models. I: Description and evaluation of the microphysical processes. *Q.J.R. Meteorol.*  
52 *Soc.*, **123**, 1227-1282.
- 53 Rotstajn, L.D., and J.E. Penner, 2001: Indirect aerosol forcing, quasi forcing, and climate response. *J.*  
54 *Climate*, **14**, 2960-2975.
- 55 Rotstajn, L.D., and Y. Liu, 2003: Sensitivity of the first indirect aerosol effect to an increase of the cloud  
56 droplet spectral dispersion with droplet number concentration. *J. Climate*, **16**, 3476-3481.

- 1 Rotstayn, L.D., B.F. Ryan, and J. Katzfey, 2000: A scheme for calculation of the liquid fraction in mixed-  
2 phase clouds in large scale models. *Mon. Wea. Rev.*, **128**, 1070-1088.
- 3 Rottman, G., 2005: The SORCE Mission. *Solar Phys.*, **230**, 7-25.
- 4 Rozanov, E.V., et al., 2002: Climate/chemistry effect of the Pinatubo volcanic eruption simulated by the  
5 UIUC stratosphere/troposphere GCM with interactive photochemistry. *J. Geophys. Res.*, **107**, 4594,  
6 doi: 10.1029/2001JD000974.
- 7 Rozanov E. V., E. M. Schlesinger, T. A. Egorova, B. Li, N. Andronova, V. A. Zubov, 2004: Atmospheric  
8 response to the observed increase of solar UV radiation from solar minimum to solar maximum  
9 simulated by the University of Illinois at Urbana-Champaign climate-chemistry model, *J. Geophys.*  
10 *Res.*, **109**, D01110, doi:10.1029/2003JD003796.
- 11 Russell, P.B., and J. Heintzenberg, 2000: An overview of the ACE-2 clear sky column closure experiment  
12 (CLEARCOLUMN). *Tellus*, **52B**, 2 463-483 doi: 10.1034/j.1600-0889.2000.00013.x.
- 13 Russell, P.B., P.V. Hobbs, and L.L. Stowe, 1999: Aerosol properties and radiative effects in the United  
14 States East Coast haze plume: An overview of the Tropospheric Aerosol Radiative Forcing  
15 Observational Experiment (TARFOX). *J. Geophys. Res.*, **104**(D2), 2213-2222.
- 16 Rypdal, K., et al., 2005: Tropospheric ozone and aerosols in climate agreements: scientific and political  
17 challenges. *Env. Science and Policy*, **8**, 29-43.
- 18 Salby, M., and P. Callaghan, 2004: Evidence of the solar cycle in the general circulation of the stratosphere.  
19 *J. Climate*, **17**, 34-46.
- 20 Santer, B.D., et al., 2004: Identification of anthropogenic climate change using a second-generation  
21 reanalysis. *J. Geophys. Res.*, **109**, D21104, doi:10.1029/2004JD005075.
- 22 Sato, M., J.E.Hansen, M.P. McCormick, and J.B. Pollack, 1993: Stratospheric aerosol optical depths, 1850-  
23 1990. *J. Geophys. Res.*, **98**(D12), 22987-22994.
- 24 Sato, M., et al., 2003: Global atmospheric black carbon inferred from AERONET. *Proc. Natl. Acad. Sci.*,  
25 **100**, 6319-6324.
- 26 Sausen, R., and U. Schumann, 2000: Estimates of the climate response to aircraft CO<sub>2</sub> and NO(x) emissions  
27 scenarios. *Clim. Change*, **44**, 27-58.
- 28 Sausen, R., K. Gierens, M. Ponater, and U. Schumann, 1998: A diagnostic study of the global distribution of  
29 contrails part I: Present day climate. *Theor. Appl. Climatol.*, **61**, 127-141.
- 30 Sausen, R., et al., 2005: Aviation radiative forcing in 2000: An update on IPCC (1999). *Meteorol. Z.*, **14**, 1-7.
- 31 Schaaf, C.B., et al., 2002: First operational BRDF, albedo nadir reflectance products from MODIS. *Remote*  
32 *Sens. Environ.*, **83**, 135-148.
- 33 Schaap M, M. van Loon, H.M. ten Brink, F.J. Dentener, and P.J.H. Builtjes, 2004: Secondary inorganic  
34 aerosol simulations for Europe with special attention to nitrate. *Atmos. Chem. Phys.*, **4**, 857-874.
- 35 Schatten, K.H., and J.A. Orosz, 1990: Solar constant secular changes. *Sol. Phys.*, **125**, 179-184.
- 36 Schmidt, G.A., et al., 2005: Present day atmospheric simulations using GISS ModelE: Comparison to in situ,  
37 satellite and reanalysis data. *J. Climate*, **19**, 153-192.
- 38 Schnaiter, M., et al., (2003): UV-VIS-NIR spectral optical properties of soot and soot-containing aerosols. *J.*  
39 *Aerosol. Sci.* **34** (10): 1421-1444.
- 40 Schoeberl, M., A. Douglass, Z. Zhu, and S. Pawson, 2003: A comparison of the lower stratospheric age  
41 spectra derived from a general circulation model and two data assimilation systems. *J. Geophys.*  
42 *Res.*, **108**, L4113, doi:10.1029/2002JD002652.
- 43 Scholes, M., and M.O. Andreae, 2000: Biogenic and pyrogenic emissions from Africa and their impact on  
44 the global atmosphere. *Ambio*, **29**, 23-29.
- 45 Schulz, M., S. Kinne, C. Textor, and S. Guibert, 2004: *AeroCom Aerosol model intercomparison.*  
46 <http://nansen.ipsl.jussieu.fr/AEROCOM/>.
- 47 Schulz, M., et al., 2006: Radiative forcing by aerosols as derived from the AeroCom present-day and pre-  
48 industrial simulations. *Atmos. Chem. Phys. Discuss.*, **6**:5095-5136.
- 49 Schumann, U., 2005: Formation, properties, and climatic effects of contrails. *C. R. Physique*, **6**, 549-565.
- 50 Schuster, G.L., O. Dubovik, B.N. Holben, and E.E. Clothiaux, 2005: Inferring black carbon content and  
51 specific absorption from Aerosol Robotic Network (AERONET) aerosol retrievals. *J. Geophys. Res.*,  
52 **110**, D10S17, doi:10.1029/2004JD004548.
- 53 Schwartz, S.E. and M.O. Andreae, 1996: Uncertainty in climate change caused by aerosols. *Science* **272**,  
54 1121-1122.
- 55 Schwartz, S.E., D.W. Harshvardhan, and C.M. Benkovitz, 2002: Influence of anthropogenic aerosol on cloud  
56 optical depth and albedo shown by satellite measurements and chemical transport modeling. *Proc.*  
57 *Natl. Acad. Sci. U.S.A.*, **99**, 1784-1789.

- 1 Sekiguchi, M., et al., 2003: A study of the direct and indirect effects of aerosols using global satellite  
2 datasets of aerosol and cloud parameters. *J. Geophys. Res.*, **108**(D22), 4699, doi:  
3 10.1029/2002JD003359.
- 4 Sellers, P.J., et al., 1996: Comparison of radiative and physiological effects of doubled atmospheric CO<sub>2</sub> on  
5 climate. *Science*, **271**, 1402-1406.
- 6 Shantz, N.C., W.R. Leaitch, and P. Caffrey, 2003: Effect of organics of low solubility on the growth rate of  
7 cloud droplets. *J. Geophys. Res.*, **108**(D5), doi: 10.1029/2002JD002540.
- 8 Sherwood, S., 2002: A microphysical connection among biomass burning, cumulus clouds, and stratospheric  
9 moisture. *Science*, **295**, 1272-1275.
- 10 Shi, G. Y., et al., 2005: Sensitivity experiments on the effects of optical properties of dust aerosols on their  
11 radiative forcing under clear sky condition. *J. Meteorol. Soc. Jpn.*, **83A**, 333-346.
- 12 Shindell, D.T., 2001: Climate and ozone response to increased stratospheric water vapor. *Geophys. Res.*  
13 *Let.*, **28**, 1551-1554.
- 14 Shindell, D.T., and G. Faluvegi, 2002: An exploration of ozone changes and their radiative forcing prior to  
15 the chlorofluorocarbon era. *Atmos. Chem. Phys.*, **2**, 363-374.
- 16 Shindell, D.T., G. Faluvegi, and N. Bell, 2003a: Preindustrial-to-presentday radiative forcing by tropospheric  
17 ozone from improved simulations with the GISS chemistry-climate GCM. *Atmos. Chem. Phys.*, **3**,  
18 1675-1702.
- 19 Shindell, D.T., G.A. Schmidt, R.L. Miller, and M. Mann, 2003b: Volcanic and solar forcing of climate  
20 change during the preindustrial era. *J. Climate*, **16**, 4094-4107.
- 21 Shindell, D.T., G.A. Schmidt, M. Mann, and G. Faluvegi, 2004: Dynamic winter climate response to large  
22 tropical volcanic eruptions since 1600. *J. Geophys. Res.*, **109**, D05104, doi:10.1029/2003JD004151.
- 23 Shindell, D.T., G. Faluvegi, N. Bell, and G. Schmidt, 2005: An emissions-based view of climate forcing by  
24 methane and tropospheric ozone. *Geophys. Res. Let.*, **32**, L04803, doi:10.1029/2004GL021900.
- 25 Shine, K.P., 2005: Comment on 'Contrails, cirrus, trends, and climate'. *J. Climate*, **18**, 2781-2782.
- 26 Shine, K.P., J. Cook, E.J. Highwood, and M.M. Joshi, 2003: An alternative to radiative forcing for  
27 estimating the relative importance of climate change mechanisms. *Geophys. Res. Let.*, **30** (20),  
28 2047, doi:10.1029/2003GL018141.
- 29 Shine, K.P., J.S. Fuglestedt, K. Hailemariam, and N. Stuber, 2005a: Alternatives to the global warming  
30 potential for comparing climate impacts of emissions of greenhouse gases. *Clim. Change*, **68**, 281-  
31 302.
- 32 Shine, K.P., T.K. Berntsen, J.S. Fuglestedt, and R. Sausen, 2005b: Scientific issues in the design of metrics  
33 for inclusion of oxides of nitrogen in global climate agreements. *Proc. Natl. Acad. Sci. U.S.A.*, **102**:  
34 15768-15773
- 35 Shine, K.P., et al., 2005c: Perfluorodecalin: global warming potential and first detection in the atmosphere.  
36 *Atmos. Env.*, **39**, 1759-1763
- 37 Simmonds, P.G., R. Derwent, A. Manning, and G. Spain, 2004: Significant growth in surface ozone at Mace  
38 Head, Ireland, 1987-2003. *Atmos. Env.*, **38**, 4769-4778.
- 39 Simpson, I.J., D.R. Blake, F.S. Rowland, and T.Y. Chen, 2002: Implications of the recent fluctuations in the  
40 growth rate of tropospheric methane. *Geophys. Res. Let.*, **29**(10), doi: 10.1029/2001GL014521.
- 41 Smith, C.A., J.D. Haigh, and R. Toumi, 2001: Radiative forcing due to trends in stratospheric water vapour.  
42 *Geophys. Res. Let.*, **28**(1), 179-182.
- 43 Smith, R.N.B., 1990: A scheme for predicting layer clouds and their water content in a general circulation  
44 model. *Q.J.R. Meteorol. Soc.*, **116**, 435-460.
- 45 Smith, S.J., and T.M.L. Wigley, 2000: Global warming potentials: 2. Accuracy. *Clim. Change*, **44**, 459-469.
- 46 Smith, S.J., E. Conception, R. Andres, and J. Lurz, 2004: *Historical sulphur dioxide emissions 1850-2000:*  
47 *methods and results*. PNNL Research Report No. PNNL-14537, Maryland, U.S., 16 p.
- 48 Smith Jr. W.L., et al., 2005: EOS terra aerosol and radiative flux validation: an overview of the  
49 Chesapeake Lighthouse and Aircraft Measurements for Satellites (CLAMS) experiment. *J. Atmos.*  
50 *Sci.*, **62**(4), 903-918, doi: 10.1175/JAS3398.1.
- 51 Sofia, S., and L.H. Li, 2001: Solar variability and climate. *J. Geophys. Res.*, **106**(A7), 12,969-12,974.
- 52 Sokolov, A., 2006: Does model sensitivity to changes in CO<sub>2</sub> provide a measure of sensitivity to the forcing  
53 of different nature. *J. Climate*, **19**, 3294-3306.
- 54 Solanki S.K., and M. Fligge, 1999: A reconstruction of total solar irradiance since 1700. *Geophys. Res. Let.*  
55 **26**(16), 2465
- 56 Solanki, S.K., I.G. Usoskin, B. Kromer, M. Schüssler, and J. Beer, 2004: Unusual activity of the Sun during  
57 recent decades compared to the previous 11,000 years. *Nature*, **431**, 1084-1087.

- 1 Solomon, S., et al., 1996: The role of aerosol variations in anthropogenic ozone depletion at northern  
2 midlatitudes. *J. Geophys. Res.*, **101**(D3), 6713-6727.
- 3 Solomon S., D.W. J. Thompson, R.W. Portmann, and S.J. Oltmans, and A.M. Thompson, 2005: On the  
4 distribution and variability of ozone in the tropical upper troposphere: Implications for tropical deep  
5 convection and chemical-dynamical coupling. *Geophys. Res. Lett.*, **32**, L23813,  
6 doi:10.1029/2005GL024323.
- 7 Soufflet, V., D. Tanre, A. Royer, and N.T. O'Neill, 1997: Remote sensing of aerosols over boreal forest and  
8 lake water from AVHRR data. *Remote Sens. Environ.*, **60**, 22-34.
- 9 Spahni, R., et al., 2005: Atmospheric methane and nitrous oxide of the late Pleistocene from Antarctic ice  
10 cores. *Science*, **310**, 1317-1321.
- 11 Spinhirne, J.D., S.P. Palm, W.D. Hart, D.L. Hlavka, and E.J. Welton, 2005: Cloud and aerosol measurements  
12 from GLAS: overview and initial results. *Geophys. Res. Lett.*, **32**, L22S03.
- 13 Spruit, H., 2000: Theory of solar irradiance variations. *Space Sci. Rev.*, **94**, 113-126.
- 14 Steinbrecht, W., H. Claude, and P. Winkler, 2004a: Enhanced upper stratospheric ozone: sign of recovery or  
15 solar cycle effect? *J. Geophys. Res.*, **109**, D020308doi:10.1029/2003JD004284.
- 16 Steinbrecht W., H. Claude, P. Winkler, 2004b: Reply to comment by D. M. Cunnold et al. on "Enhanced  
17 upper stratospheric ozone: Sign of recovery or solar cycle effect?". *J. Geophys. Res.*, **109**, D14306,  
18 doi:10.1029/2004JD004948.
- 19 Stenchikov, G., K. Hamilton, A. Robock, V. Ramaswamy, and M. D. Schwarzkopf, 2004: Arctic oscillation  
20 response to the 1991 Pinatubo eruption in the SKYHI GCM with a realistic quasi-biennial  
21 oscillation. *J. Geophys. Res.*, **109**, D03112, doi:10.1029/2003JD003699.
- 22 Stenchikov, G., et al., 2006: Arctic Oscillation response to Volcanic Eruptions in the IPCC AR4 Climate  
23 Models. *J. Geophys. Res.*, **111**, D07107, doi:10.1029/2005JD006286.
- 24 Stenchikov, G.L., et al., 2002: Arctic oscillation response to the 1991 Mount Pinatubo eruption: effects of  
25 volcanic aerosols and ozone depletion. *J. Geophys. Res.*, **107**(D24), 4803,  
26 doi:10.1029/2002JD002090.
- 27 Stenchikov, G.L., et al., 1998: Radioactive forcing from the 1991 Mount Pinatubo volcanic eruption. *J.*  
28 *Geophys. Res.*, **103**(D12), 13,837-13,857.
- 29 Stern, D.I., 2005: Global sulfur emissions from 1850 to 2000. *Chemosphere*, **58**, 163-175.
- 30 Stevenson, D.S., R.M. Doherty, M.G. Sanderson, W.J. Collins, C.E. Johnson, and R.G. Derwent, 2004:  
31 Radiative forcing from aircraft NO<sub>x</sub> emissions: mechanisms and seasonal dependence. *J. Geophys.*  
32 *Res.*, **109**, D17307, doi:10.1029/2004JD004759.
- 33 Stier, P., J. et al., 2005: The aerosol-climate model ECHAM5-HAM. *Atmos. Chem. Phys.*, **5**, 1125-1156.
- 34 Stier, P., J. H. Seinfeld, S. Kinne, J. Feichter, and O. Boucher. 2006a. The Impact of non-absorbing  
35 anthropogenic aerosols on atmospheric absorption. *J. Geophys. Res.* (in press).
- 36 Stier, P., J. Feichter, S. Kloster, E. Vignati, and J. Wilson. 2006b. Emission-induced nonlinearities in the  
37 global aerosol system: results from the ECHAM5-HAM aerosol-climate model. *J. of Climate*, **19**,  
38 3845-3862
- 39 Stordal, F., et al., 2005: Is there a trend in cirrus cloud cover due to aircraft traffic? *Atmos. Chem. Phys.*, **5**,  
40 2155-2162.
- 41 Stothers, R., 2001a: Major optical depth perturbations to the stratosphere from volcanic eruptions: Stellar  
42 extinction period, 1961-1978. *J. Geophys. Res.*, **106**(D3), 2993-3003.
- 43 Stothers, R., 2001b: A chronology of annual mean radii of stratospheric aerosol from volcanic eruptions  
44 during the twentieth century as derived from ground-based spectral extinction measurements. *J.*  
45 *Geophys. Res.*, **106**(D23), 32,043-32,049.
- 46 Stott P.A., G.S. Jones, and J.F.B. Mitchell, 2003: Do models underestimate the solar contribution to recent  
47 climate change? *J. Climate*, **16**, 4079-4093
- 48 Streets, D.G., S. Gupta, S.T. Waldhoff, M.Q.Wang, T.C. Bond, and Y. Bo, 2001: Black carbon emissions in  
49 China. *Atmos. Environ.*, **35**, 4281- 4296.
- 50 Streets, D.G., et al., 2003): An inventory of gaseous and primary aerosol emissions in Asia in the year 2000.  
51 *J. Geophys. Res.*, **108**(D21), 8809, doi:10.1029/2002JD003093.
- 52 Strugnell, N.C., W. Lucht, and C. Schaaf, 2001: A global albedo data set derived from AVHRR data for use  
53 in climate simulations. *Geophys. Res. Lett.*, **28**(1), 191-194.
- 54 Stuber, N., R. Sausen, and M. Ponater, 2001a: Stratosphere adjusted radiative forcing calculations in a  
55 comprehensive climate model. *Theor. Appl. Climatol.*, **68**, 125-135.
- 56 Stuber, N., M. Ponater, and R. Sausen, 2001b: Is the climate sensitivity to ozone perturbations enhanced by  
57 stratospheric water vapor feedback? *Geophys. Res. Lett.*, **28**(15), 2887-2890.

- 1 Stuber, N., M. Ponater, and R. Sausen, 2005: Why radiative forcing might fail as a predictor of climate  
2 change. *Clim. Dyn.*, **24**, 497-510.
- 3 Stubenrauch, C.J. and U. Schumann, 2005: Impact of air traffic on cirrus coverage. *Geophys. Res. Lett.*, **32**,  
4 L14813, doi:10.1029/2005GL022707.
- 5 Sun, B., and R.S. Bradley, 2002: Solar influences on cosmic rays and cloud formation: a re-assessment. *J.*  
6 *Geophys. Res.*, **107**(D14), doi:10.1029/2001JD000560.
- 7 Sundqvist, H., 1978: A parameterization scheme for non-convective condensation including prediction of  
8 cloud water content. *Q. J. R. Meteorol. Soc.*, **104**, 677-690.
- 9 Sundqvist, H., E. Berge, and J.E. Kristjánsson, 1989: Condensation and cloud parameterization studies with  
10 a mesoscale numerical weather prediction model. *Mon. Wea. Rev.*, **117**, 1641-1657.
- 11 Suzuki, K., et al., 2004: A study of the aerosol effect on a cloud field with simultaneous use of GCM  
12 modeling and satellite observation. *J. Atmos. Sci.*, **61**, 179-194.
- 13 Svalgaard, L., E.W. Cliver, and P. Le Sager, 2004: IHV: A new long-term geomagnetic index. *Adv. Space*  
14 *Res.*, **34**, 436-439.
- 15 Swap, R.J., et al., 2003: Africa burning: a thematic analysis of the Southern African Regional Science  
16 Initiative (SAFARI 2000). *J. Geophys. Res.*, **108**(D13), 8465, doi:10.1029/2003JD003747.
- 17 Swap, R.J., et al., 2002: The Southern African Regional Science Initiative (SAFARI 2000): overview of the  
18 dry season field campaign. *S. Afr. J. Sci.*, **98**, 125-130.
- 19 Tabazadeh, A., K. Drdla, M.R. Schoeberl, P. Hamill, and O.B. Toon, 2002: Arctic "ozone hole" in cold  
20 volcanic stratosphere. *Proc. Natl. Acad. Sci.*, **99**, 2609-2612.
- 21 Takemura, T., T. Nakajima, T. Nozawa, and K. Aoki, 2001: Simulation of Future Aerosol Distribution,  
22 Radiative Forcing, and Long-Range Transport in East Asia. *J. Meteorol. Soc. Jpn.*, **2**, 79, 1139-1155.
- 23 Takemura, T., T. Nozawa, S. Emori, T.Y. Nakajima, and T. Nakajima, 2005: Simulation of climate response  
24 to aerosol direct and indirect effects with aerosol transport-radiation model. *J. Geophys. Res.*, **110**,  
25 D02202 doi:10.1029/2004JD005029.
- 26 Takemura, T., et al., 2000: Global three-dimensional simulation of aerosol optical thickness distribution of  
27 various origins. *J. Geophys. Res.*, **105**(D14), 17853-17874.
- 28 Tang, I.N., 1997: Thermodynamic and optical properties of mixed-salt aerosols of atmospheric importance.  
29 *J. Geophys. Res.*, **102**(D2), 1883-1893.
- 30 Tang, I.N., K.H. Fung, D.G. Imre, and H.R. Munkelwitz, 1995: Phase Transformation and Metastability of  
31 Hygroscopic Microparticles. *Aerosol Sci. Tech.*, **23**, 443.
- 32 Tanré, D., Y.J. Kaufman, M. Herman, S. Mattoo, 1997: Remote sensing of aerosol properties over oceans  
33 using the MODIS/EOS spectral radiances. *J. Geophys. Res.*, **102**(D14), 16971-16988.
- 34 Tanré, D., et al., 2003: Measurement and modeling of the Saharan dust radiative impact: overview of the  
35 SaHARan Dust Experiment (SHADE). *J. Geophys. Res.*, **108**(D18), doi:10.1029/2002JD003273.
- 36 Tarasick, D.W., V. Fioletov, D. Wardle, J. Kerr, and J. Davies, 2005: Changes in the vertical distribution of  
37 ozone over Canada from ozonesondes: 1980-2001. *J. Geophys. Res.*, **110**,  
38 D02304doi:10.1029/2004JD004643.
- 39 Tegen, I., and I. Fung, 1995: Contribution to the atmospheric mineral aerosol load from land surface  
40 modification. *J. Geophys. Res.*, **100**, 18,707.
- 41 Tegen, I., A.A. Lacis, and I. Fung, 1996: The influence on climate forcing of mineral aerosols from disturbed  
42 soils. *Nature*, **380**, 419-421.
- 43 Tegen, I., M. Werner, S.P. Harrison, and K.E. Kohfeld, 2004: Relative importance of climate and land use in  
44 determining present and future global soil dust emission. *Geophys. Res. Lett.*, **31**, L05105,  
45 doi:10.1029/2003GL019216.
- 46 Tegen, I., M. Werner, S.P. Harrison, and K.E. Kohfeld, 2005: Reply to comment by N. M. Mahowald et al.  
47 on "Relative importance of climate and land use in determining present and future global soil dust  
48 emission,". *Geophys. Res. Lett.*, **32**, doi:10.1029/2004GL021560.
- 49 Tett, S.F.B., et al., 2002: Estimation of natural and anthropogenic contributions to twentieth century  
50 temperature change. *J. Geophys. Res.*, **107**(D16), 4306, doi:10.1029/2000JD000028.
- 51 Textor, C., et al. 2006: AeroCom: The status quo of global aerosol modelling. *Atmos. Chem. Phys.* **6**, 1777-  
52 1813.
- 53 Tie, X. X., G.P. Brasseur, B. Breiglib and C. Granier, 1994: Two dimensional simulation of Pinatubo aerosol  
54 and its effect on stratospheric chemistry. *J. Geophys. Res.*, **99**(D10), 20545-20562.
- 55 Thompson, A.M., J. Witte, R. Hudson, H. Guo, J. Herman, and M. Fujiwara, 2001: Tropical tropospheric  
56 ozone and biomass burning. *Science*, **291**, 2128-2132.

- 1 Thompson, T.M., et al., 2004: Halocarbons and other Atmospheric Trace Species. In: Climate Monitoring  
2 and Diagnostics Laboratory, Summary Report No. 27 [Schnell, R.C., A.-M. Buggle, and R.M.  
3 Rosson (eds.)], NOAA CMDL, Boulder, CO, pp. 115-135.
- 4 Thomason, L., and T. Peter, 2006: Assessment of Stratospheric Aerosol Properties (ASAP): Report on the  
5 Assessment Kick-Off Workshop, Paris, France, 4-6 November 2001. SPARC Report No. 4 ; WCRP-  
6 124, WMO/TD No. 1295, [http://www.aero.jussieu.fr/~sparc/News18/18\\_Thomason.html](http://www.aero.jussieu.fr/~sparc/News18/18_Thomason.html).
- 7 Timmreck, C., and M. Schulz, 2004: Significant dust simulation differences in nudged and climatological  
8 operation mode of the AGCM ECHAM. *J. Geophys. Res.*, **109**, D13202,  
9 doi:10.1029/2003JD004381.
- 10 Timmreck, C., H.-F. Graf, and B. Steil, 2003: Aerosol chemistry interactions after the Mt. Pinatubo  
11 Eruption. In: *Volcanism and the Earth's Atmosphere*. [Robock, A. and C. Oppenheimer (eds.)].  
12 Geophysical Monograph 139, American Geophysical Union, Washington, D.C., pp. 227-236.
- 13 Tol, R.S.J., 2002: Estimates of the Damage Costs of Climate Change, Part II. Dynamic Estimates. *Environ.*  
14 *Resour. Econ.*, **21**, 135-160.
- 15 Torres, O., et al., 2002: A Long-Term Record of Aerosol Optical Depth from TOMS Observations and  
16 Comparison to AERONET Measurements. *J. Atmos. Sci.*, **59**, 398-413.
- 17 Travis, D.J., A.M. Carleton, and R.G. Lauritsen, 2002: Contrails reduce daily temperature range. *Nature*,  
18 **418**, 601-602.
- 19 Travis, D.J., A.M. Carleton, and R.G. Lauritsen, 2004: Regional variations in U.S. diurnal temperature range  
20 for the 11-14 September 2001 aircraft groundings: evidence of jet contrail influence on climate. *J.*  
21 *Climate*, **17**, 1123-1134.
- 22 Tripoli, G.J., and W.R. Cotton, 1980: A numerical investigation of several factors contributing to the  
23 observed variable intensity of deep convection over South Florida. *J. Appl. Meteorol.*, **19**, 1037-  
24 1063.
- 25 Twohy, C. H., et al., 2005: Evaluation of the aerosol indirect effect in marine stratocumulus clouds: droplet  
26 number, size, liquid water path, and radiative impact. *J. Geophys. Res.*, **110**, D08203,  
27 doi:10.1029/2004JD005116.
- 28 Twomey, S.A., 1977: The influence of pollution on the shortwave albedo of clouds. *J. Atmos. Sci.*, **34**, 1149-  
29 1152.
- 30 Udelhofen, P.M., and R.D. Cess, 2001: Cloud cover variations over the United States: An influence of  
31 cosmic rays or solar variability? *Geophys. Res. Lett.*, **28**(13), 2617-2620.
- 32 Usoskin, I.G., N. Marsh, G.A. Kovaltsov, K. Mursula, and O.G. Gladysheva, 2004: Latitudinal dependence  
33 of low cloud amount on cosmic ray induced ionization. *Geophys. Res. Lett.*, **31**, L16109.
- 34 Van Aardenne, J.A., F.J. Dentener, J.G.J. Olivier, C.G.M. Klein Goldewijk, and J. Lelieveld, 2001: A 1 x 1  
35 degree resolution dataset of historical anthropogenic trace gas emissions for the period 1890-1990.  
36 *Global Biogeochem. Cycles*, **15**, 909-928.
- 37 van der Werf, et al., 2004: Continental-scale partitioning of fire emissions during the 1997 to 2001 El  
38 Nino/La Nina period. *Science*, **303**, 73-76.
- 39 Van Dorland, R., F.J. Dentener, and J. Lelieveld, 1997: Radiative forcing due to tropospheric ozone and  
40 sulfate aerosols. *J. Geophys. Res.*, **102**(D23), 28,079-28,100.
- 41 van Loon, H., and D.J. Shea, 2000: The global 11-year solar signal in July-August. *Geophys. Res. Lett.*,  
42 **27**(18), 2965-2968.
- 43 Veefkind, J. P., G. de Leeuw, and P.A. Durkee, 1998: Retrieval of aerosol optical depth over land using two-  
44 angle view satellite radiometry. *Geophys. Res. Lett.* **25**(16), 3135-3138.
- 45 Velders, et al., 2005: Chemical and radiative effects of halocarbons and their replacement compounds. In:  
46 *Special Report on Safeguarding the Ozone Layer and Global Climate System*. IPCC/TEAP,  
47 Cambridge U.K., pp. 133-180.
- 48 Vestreng, V. , M. Adams, and J. Goodwin, 2004: *Inventory Review 2004: Emission Data Reported to*  
49 *CLRTAP and the NEC Directive*. EMEP/EEA Joint Review Report, Norway, 120 p.
- 50 Walter, B.P., M. Heimann, and E. Matthews, 2001a: Modeling modern methane emissions from natural  
51 wetlands 2. Interannual variations 1982-1993. *J. Geophys. Res.*, **106**(D24), 34207-34220.
- 52 Walter, B.P., M. Heimann, and E. Matthews, 2001b: Modeling modern methane emissions from natural  
53 wetlands 1. Model description and results. *J. Geophys. Res.*, **106**(D24), 34189-34206.
- 54 Wang, C., 2004: A modeling study on the climate impacts of black carbon aerosols. *J. Geophys. Res.*, **109**,  
55 D03106, doi:10.1029/2003JD004084.
- 56 Wang, H.J., D.M. Cunnold, L.W. Thomas, J.M. Zawodny, and G.E. Bodeker, 2002: Assessment of SAGE  
57 version 6.1 ozone data quality. *J. Geophysics. Res.*, **107**(D23), 4691, doi:10.1029/2002JD002418.

- 1 Wang, M.H., K.D. Knobelspiesse, and C.R. McClain, 2005: Study of the Sea-Viewing Wide Field-of-View  
2 Sensor (SeaWiFS) aerosol optical property data over ocean in combination with the ocean color  
3 products. *J. Geophys. Res.*, **110**, D10S06.
- 4 Wang, W.-C., M.P. Dudek, X.-Z. Liang, and J.T. Kiehl, 1991: Inadequacy of effective CO<sub>2</sub> as a proxy in  
5 simulating the greenhouse effect of other radiatively active gases. *Nature*, **350**, 573-577.
- 6 Wang, W.-C., M.P. Dudek, and X.-Z. Liang, 1992: Inadequacy of effective CO<sub>2</sub> as a proxy in assessing the  
7 regional climate change due to other radiatively active gases. *Geophys. Res. Lett.*, **19**, 1375-1378.
- 8 Wang, Y.M., J.L. Lean, and N.R. Sheeley, 2005: Modeling the sun's magnetic field and irradiance since  
9 1713. *Astrophys. J.*, **625**, 522-538.
- 10 Warner, J., and S.A. Twomey, 1967: The production and cloud nuclei by cane fires and the effect on cloud  
11 droplet concentration. *J. Atmos. Sci.*, **24**, 704-706.
- 12 Warwick, N.J., S. Bekki, K.S. Law, E.G., Nisbet, and J.A. Pyle, 2002: The impact of meteorology on the  
13 interannual growth rate of atmospheric methane. *Geophys. Res. Lett.*, **29**(26), doi:  
14 10.1029/2002/GL015282.
- 15 Weatherhead, E. C. and S. B. Andersen, 2006: The search for signs of recovery of the ozone layer. *Nature*,  
16 **441**, 39-45.
- 17 Welton, E.J., J.R. Campbell, J.D. Spinhirne, and V.S. Scott, 2001: Global monitoring of clouds and aerosols  
18 using a network of micro-pulse lidar systems. In: *Lidar Remote Sensing for Industry and*  
19 *Environmental Monitoring*. [Singh, U.N., T. Itabe, and N. Sugimoto (eds.)]. SPIE, Bellingham, Wash.,  
20 pp. 151-158.
- 21 Wennberg, P.O., S. Peacock, J.T. Randerson, and R. Bleck, 2004: Recent changes in the air-sea gas  
22 exchange of methyl chloroform. *Geophys. Res. Lett.*, **31**, L16112, doi:10.1029/2004GL020476.
- 23 Westerling, A.L., H.G. Hidalgo, D.R. Cayan, T.W. Swetnam, 2006: Warming and earlier spring increase  
24 western U.S. forest wildfire activity. *Science*, **313**, 940-943.
- 25 White, W.B., M.D. Dettinger, and D.R. Cayan, 2003: Sources of global warming of the upper ocean on  
26 decadal period scales. *J. Geophys. Res.*, **108**(C8), doi:10.1029/2002JC001396.
- 27 Wild, O., M.J. Prather, and H. Akimoto, 2001: Indirect long-term global radiative cooling from NO<sub>x</sub>  
28 emissions. *Geophys. Res. Lett.*, **28**(9), 1719-1722.
- 29 Williams, K.D., C.A. Senior, and J.F.B. Mitchell, 2001a: Transient climate change in the Hadley Centre  
30 models: the role of physical processes. *J. Climate*, **14**, 2659-2674.
- 31 Williams, K.D., A. Jones, D.L. Roberts, C.A. Senior, and M.J. Woodage, 2001b: The response of the climate  
32 system to the indirect effects of anthropogenic sulfate aerosols. *Clim. Dyn.*, **17**, 846-856.
- 33 Willson, R.C., and A.V. Mordvinov, 2003: Secular total solar irradiance trend during solar cycles 21-23.  
34 *Geophys. Res. Lett.*, **30**(5), 3-6.
- 35 Wilson, D.R., and S.P. Ballard, 1999: A microphysically based precipitation scheme for the UK  
36 Meteorological Office Unified Model. *Q.J.R. Meteorol. Soc.*, **125**, 1607-1636.
- 37 Wilson, M.F., and A. Henderson-Sellers, 1985: A global archive of land cover and soils data for use in  
38 general-circulation climate models. *J. Climatol*, **5**, 119-143.
- 39 WMO, 1986: Atmospheric Ozone 1985. Global Ozone Research and Monitoring Project No.16, World  
40 Meteorological Organisation, Geneva, 3 v.
- 41 WMO, 2003: Scientific Assessment of Ozone Depletion: 2002. Global Ozone Research and Monitoring  
42 Project. Report number 47, World Meteorological Organization, Geneva, 498 p.
- 43 Wong, J., and Z. Li, 2002: Retrieval of optical depth for heavy smoke aerosol plumes: uncertainties and  
44 sensitivities to the optical properties. *J. Atmos. Sci.*, **59**, 250-261.
- 45 Wong, S., W.-C. Wang, I. Isaksen, T. Berntsen, and J. Sundet, 2004: A global climate-chemistry model  
46 study of present-day tropospheric chemistry and radiative forcing from changes in tropospheric O<sub>3</sub>  
47 since the preindustrial period. *J. Geophys. Res.*, **109**, D11309, doi:10.1029/2003JD003998.
- 48 Woods, T.N., et al., 1996: Validation of the UARS solar ultraviolet irradiances: comparison with the ATLAS  
49 1 and 2 measurements. *J. Geophys. Res.*, **101**(D6), 9541-9569.
- 50 Xiong, J.Q., M.H. Zhong, C.P. Fang, L.C. Chen, and M. Lippmann, 1998: Influence of organic films on the  
51 hygroscopicity of ultrafine sulfuric acid aerosol. *Environ. Sci. Technol.*, **32**, 3536-3541.
- 52 Xue, H and G. Feingold, 2006: large eddy simulations of trade-wind cumuli: investigation of aerosol indirect  
53 effects. *J. Atmos. Sci.*, **63**, 1605-1622.
- 54 Yang, F., and M. Schlesinger, 2001: Identification and separation of Mount Pinatubo and El Nino-Southern  
55 Oscillation land surface temperature anomalies. *J. Geophys. Res.*, **106**(D14), 14,757-14,770.
- 56 Yang, F., and M. Schlesinger, 2002: On the surface and atmospheric temperature changes following the 1991  
57 Pinatubo volcanic eruption: a GCM study. *J. Geophys. Res.*, **107**(D8), doi:10.1029/2001JD000373.

- 1 Yokouchi, et al., 2005: Estimates of ratios of anthropogenic halocarbon emissions from Japan based on  
2 aircraft monitoring over Sagami Bay, Japan. *J. Geophys. Res.*, **110**, D06301,  
3 doi:10.1029/2004JD005320.
- 4 Yoshioka, M., N. Mahowald, J.L. Dufresne, and C. Luo, 2005: Simulation of absorbing aerosol indices for  
5 African dust. *J. Geophys. Res.*, **110**, D18S17, doi:10.1029/2004JD005276.
- 6 Yvon-Lewis, S.A., and J.H. Butler, 2002: Effect of oceanic uptake on atmospheric lifetimes of selected trace  
7 gases. *J. Geophys. Res.*, **107**(D20), 4414, doi:10.1029/2001JD001267
- 8 Yu, H., et al., 2003: Annual cycle of global distributions of aerosol optical depth from integration of MODIS  
9 retrievals and GOCART model simulations. *J. Geophys. Res.*, **108**(D3), 4128,  
10 doi:10.1029/2002JD002717.
- 11 Yu, H., et al., 2006: A review of measurement-based assessments of the aerosol direct radiative effect and  
12 forcing. *Atmos. Chem. Phys.*, **6**, 613-666.
- 13 Zender, C.S., 2004: Quantifying mineral dust mass budgets: terminology, constraints, and current estimates.  
14 *EOS*, **85**, 509-512.
- 15 Zerefos, C.S., et al., 2003: Evidence of impact of aviation on cirrus cloud formation. *Atmos. Chem. Phys.*, **3**,  
16 1633-1644.
- 17 Zhang, J.P., Z. Yang, D.J. Wang, and X.B. Zhang, 2002: Climate change and causes in the Yuanmou dry-hot  
18 valley of Yunnan China. *J. Arid Environ.*, **51**, 153-162.
- 19 Zhang, J., and S. Christopher, 2003: Longwave radiative forcing of Saharan dust aerosols estimated from  
20 MODIS, MISR and CERES observations on Terra. *Geophys. Res. Lett.*, **30**(23),  
21 doi:10.1029/2003GL018479.
- 22 Zhang, J., S.A. Christopher, L.A. Remer, and Y.J. Kaufman, 2005: Shortwave aerosol radiative forcing over  
23 cloud-free oceans from Terra: 2. Seasonal and global distributions. *J. Geophys. Res.*, **110**, D10S24,  
24 doi:10.1029/2004JD005009.
- 25 Zhao, M., A. Pitman, and T.N. Chase, 2001: The impacts of land cover change on the atmospheric  
26 circulation. *Clim. Dyn.*, **17**, 467-477.
- 27 Zhao, T. X. P., I. Laszlo, P. Minnis, and L. Remer, 2005: Comparison and analysis of two aerosol retrievals  
28 over the ocean in the terra/clouds and the earth's radiant energy system: moderate resolution imaging  
29 spectroradiometer single scanner footprint data: 1. Global evaluation. *J. Geophys. Res.*, **110**,  
30 D21208.
- 31 Zhou, L.M., et al., 2001: Variations in northern vegetation activity inferred from satellite data of vegetation  
32 index during 1981 to 1999. *J. Geophys. Res.*, **106** (D17): 20069-20083.
- 33 Zhou M., H. Yu, R.E. Dickinson, O. Dubovik, and B.N. Holben, 2005: A normalized description of the  
34 direct effect of key aerosol types on solar radiation as estimated from aerosol robotic network  
35 aerosols and moderate resolution Imaging spectroradiometer albedos. *J. Geophys. Res.*, **110**,  
36 D19202, doi:10.1029/2005JD005909.
- 37  
38

## Frequently Asked 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 Earth's atmosphere in the amounts of greenhouse gases, aerosols (small particles), and cloudiness. The largest known contribution comes from the burning of fossil fuels, which releases carbon dioxide gas to the atmosphere. Greenhouse gases and aerosols affect climate by altering incoming solar radiation and outgoing infrared (thermal) radiation that are part of Earth's energy balance. Changing the atmospheric abundance or properties of these gases and particles can lead to a warming or cooling of the climate system. Since the start of the industrial era (about 1750), the overall effect of human activities on climate has been a warming influence. The human impact on climate during this era greatly exceeds that due to known changes in natural processes, such as solar changes and volcanic eruptions.

### Greenhouse-Gases

Human activities result in emissions of four principal greenhouse gases: carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O), 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 FAQ 2.1, Figure 1). All of these increases are 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 releases carbon dioxide and reduces its uptake by plants. Carbon dioxide is also released in natural processes such as the decay of plant matter.
- *Methane* has increased as a result of human activities related to agriculture, natural gas distribution, and landfills. Methane is also released in natural processes that occur, for example, in wetlands. Methane concentrations are not currently increasing in the atmosphere because growth rates decreased over the last two decades.
- *Nitrous oxide* is also emitted by human activities such as fertilizer use and fossil fuel burning. Natural processes in soils and the oceans also release nitrous oxide.

[INSERT FAQ 2.1, FIGURE 1 HERE]

- *Halocarbon gas* concentrations have increased primarily due to human activities. Natural processes are also a small source. Principal halocarbons include the chlorofluorocarbons (e.g., CFC-11 and CFC-12), which were used extensively as refrigeration agents and in other industrial processes before their presence in the atmosphere was found to cause stratospheric ozone depletion. The abundance of chlorofluorocarbon gases is decreasing as a result of international regulation designed to protect the ozone layer.
- *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, hydrocarbons, and nitrogen oxide, which chemically react to produce ozone. As mentioned above, halocarbons released in human activities destroy ozone in the stratosphere and have caused the ozone hole over Antarctica.
- *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 have the potential to affect water vapour substantially by changing climate. For example, a warmer atmosphere contains more water vapour. Human activities also influence water vapour through methane emissions, because methane undergoes chemical destruction in the stratosphere, producing a small amount of water vapour.
- *Aerosols* are small particles present in the atmosphere with widely varying size, concentration, and chemical composition. Some aerosols are emitted directly into the atmosphere while others are formed from emitted compounds. Aerosols contain both naturally occurring compounds and those emitted as a result of human activities. Fossil fuel and biomass burning have increased aerosols containing sulphur compounds, organic compounds, and black carbon (soot). Human activities such as surface mining and industrial processes have increased dust in the atmosphere. Natural aerosols include mineral dust

1 released from the surface, sea salt aerosols, biogenic emissions from the land and oceans, and sulphate  
2 and dust aerosols produced by volcanic eruptions.

3  
4 [START OF FAQ 2.1, BOX 1]

5  
6 **What is radiative forcing?** The influence of a factor that can cause climate change, such as a greenhouse gas,  
7 is often evaluated in terms of its radiative forcing. Radiative forcing is a measure of how the energy balance  
8 of the Earth-atmosphere system is influenced when factors that affect climate are altered. The word *radiative*  
9 arises because these factors change the balance between incoming solar radiation and outgoing infrared  
10 radiation involving the Earth's surface and its atmosphere. The term *forcing* is used to indicate that Earth's  
11 radiative balance is being pushed or strained away from its normal state.

12  
13 Radiative forcing is usually quantified as the 'rate of energy change per unit area of the globe as measured at  
14 the top of the atmosphere', and is expressed in units of 'Watts per square meter' (see FAQ 2.1, Figure 2).  
15 When radiative forcing from a factor or group of factors is evaluated as *positive*, the energy of the Earth-  
16 atmosphere system will ultimately *increase*, leading to a *warming* of the system. In contrast, for a *negative*  
17 radiative forcing, the energy will ultimately *decrease*, leading to a *cooling* of the system. Important  
18 challenges for climate scientists are to identify all the factors that affect climate and the mechanisms by  
19 which they exert a forcing, to quantify the radiative forcing of each factor, and to evaluate the total radiative  
20 forcing from the group of factors.

21  
22 [END OF FAQ 2.1, BOX 1]

#### 23 24 *Radiative Forcing of factors affected by human activities*

25 The contributions to radiative forcing from some of the factors influenced by human activities are shown in  
26 FAQ 2.1, Figure 2. The values reflect the total forcing relative to the start of the industrial era (about 1750).  
27 The forcings for all greenhouse gas increases, which are the best understood of those due to human activities,  
28 are positive because each gas absorbs outgoing infrared radiation in the atmosphere. Amongst the  
29 greenhouse gases, carbon dioxide increases have caused the largest forcing over this period. Tropospheric  
30 ozone increases have also contributed to warming, while stratospheric ozone decreases have contributed to  
31 cooling.

32  
33 Aerosol particles influence radiative forcing *directly* through reflection and absorption of solar and infrared  
34 radiation in the atmosphere. Some aerosols cause a positive forcing while others cause a negative forcing.  
35 The direct radiative forcing summed over all aerosol types is negative. Aerosols also cause a negative  
36 radiative forcing *indirectly* through the changes they cause in cloud properties.

37  
38 Human activities since the industrial era have altered the nature of land cover over the globe, principally  
39 through changes in croplands, pastures, and forests. They have also modified the reflective properties of ice  
40 and snow. Overall, it is likely that more solar radiation is now being reflected from Earth's surface as a result  
41 of human activities. This change results in a negative forcing.

42  
43 [INSERT FAQ 2.1, FIGURE 2 HERE]

44  
45 Aircraft produce persistent linear trails of condensation ('contrails') in regions that have suitably low  
46 temperatures and high humidity. Contrails are a form of cirrus cloud that reflects solar radiation and absorbs  
47 infrared radiation. Linear contrails from global aircraft operations have increased Earth's cloudiness and are  
48 estimated to cause a small positive radiative forcing.

#### 49 50 *Radiative Forcing from Natural Changes*

51 The largest changes in natural forcings in the industrial era are solar changes and explosive volcanic  
52 eruptions. Solar output has increased gradually in the industrial era, causing a small positive radiative forcing  
53 (see FAQ 2.1, Figure 2). This is in addition to the cyclic changes in solar radiation that follow an 11-year  
54 cycle. Solar energy directly heats the climate system and can also affect the atmospheric abundance of some  
55 greenhouse gases, such as stratospheric ozone. Explosive volcanic eruptions can create a short-lived (2 to 3  
56 years) negative forcing through the temporary increases that occur in sulphate aerosol in the stratosphere.

1 The stratosphere is currently free of volcanic aerosol, since the last major eruption was in 1991 (Mt.  
2 Pinatubo).

3  
4 The differences in radiative forcing estimates between the present day and the start of the industrial era for  
5 solar irradiance changes and volcanoes are both small compared to the differences in radiative forcing  
6 estimated to have resulted from human activities. As a result, in today's atmosphere, the radiative forcing  
7 from human activities is much more important for current and future climate change than the estimated  
8 radiative forcing from changes in natural processes.  
9

1 **Tables**

2  
3 **Table 2.1.** Present day concentrations and RF for the measured LLGHGs. The changes since 1998 (the time  
4 of the TAR estimates) are also shown.  
5

Species	Concentrations and their changes		Radiative Forcing	
	2005 (ppt)	2005–1998 (ppt)	2005 (W m <sup>-2</sup> )	Change since 1998
<b>CO<sub>2</sub></b>	<b>379 ± 0.65 ppm</b>	<b>+13 ppm</b>	<b>1.66</b>	<b>+13%</b>
<b>CH<sub>4</sub></b>	<b>1774 ± 1.8 ppb</b>	<b>+11 ppb</b>	<b>0.48</b>	<b>-</b>
<b>N<sub>2</sub>O</b>	<b>319 ± 0.12 ppb</b>	<b>+5 ppb</b>	<b>0.16</b>	<b>+11%</b>
CFC-11	251 ± 0.36	-13	0.063	-5%
CFC-12	538 ± 0.18	+4	0.17	+1%
CFC-113	79 ± 0.064	-4	0.024	-5%
HCFC-22	169 ± 1.0	+38	0.033	+29%
HCFC-141b	18 ± 0.068	+9	0.0025	+93%
HCFC-142b	15 ± 0.13	+6	0.0031	+57%
CH <sub>3</sub> CCl <sub>3</sub>	19 ± 0.47	-47	0.0011	-72%
CCl <sub>4</sub>	93 ± 0.17	-7	0.012	-7%
HFC-125	3.7 ± 0.10 <sup>a</sup>	+2.6 <sup>f</sup>	0.0009	+234%
HFC-134a	35 ± 0.73	+27	0.0055	+349%
HFC-152a	3.9 ± 0.11 <sup>a</sup>	+2.4 <sup>f</sup>	0.0004	+151%
HFC-23	18 ± 0.12 <sup>b,e</sup>	+4	0.0033	+29%
SF <sub>6</sub>	5.6 ± 0.038 <sup>c</sup>	+1.5	0.0029	+36%
CF <sub>4</sub> (PFC-14)	74 ± 1.6 <sup>d</sup>	-	0.0034	-
C <sub>2</sub> F <sub>6</sub> (PFC-116)	2.9 ± 0.025 <sup>b,e</sup>	+0.5	0.0008	+22%
<b>CFCs Total</b>			<b>0.268</b>	<b>-1%</b>
<b>HCFC Total</b>			<b>0.039</b>	<b>+33%</b>
<b>Montreal Gases</b>			<b>0.320</b>	<b>-1%</b>
<b>Other Kyoto Gases as HFCs+PFCs+SF<sub>6</sub></b>			<b>0.017</b>	<b>+69%</b>
<b>Halocarbons</b>			<b>0.337</b>	<b>+1%</b>
<b>Total LLGHGs</b>			<b>2.63</b>	<b>+9%</b>

6 Notes:

7 Mixing ratio errors are 90% confidence ranges of combined 2005 data, including intra-annual standard deviation,  
8 measurement and global averaging uncertainty. Standard deviations have been multiplied by 1.645 to obtain an estimate  
9 of the 90% confidence range; this assumes normal distributions

10 Percent changes are calculated relative to 1998 reference.

11 90% confidence ranges in RF range are not shown but are approximately 10%. This confidence range is almost entirely  
12 due to radiative transfer assumptions, therefore trends remain valid when quoted to higher accuracies.

13 Higher precision data for totals are used and impact rounding of the values. CO<sub>2</sub> are combined measurements from the  
14 ESRL and SIO networks (see Section 2.3.1).

15 CH<sub>4</sub> measurements are combined data from the ESRL and AGAGE networks (see Section 2.3.2).

16 Halocarbon measurements are average of ESRL and AGAGE networks. UEA and PSU measurements were also used  
17 (see Section 2.3.3). CFC total includes a small 0.009 W m<sup>-2</sup> RF from CFC-13, CFC-114, CFC-115 and the halons, as  
18 measurements of these were not updated.

19 Preindustrial values are zero except for: CO<sub>2</sub> (278 ppm), CH<sub>4</sub> (715 ppb; 700 ppb used in the TAR), N<sub>2</sub>O (270 ppb), CF<sub>4</sub>  
20 (40 ppt)

21 Radiative efficiencies are from Table 2.14

22 (a) Data available from AGAGE network only

23 (b) Data from UEA only

24 (c) Data from CMDL only

25 (d) 1997 data from PSU (Khalil et al., 2003), (not updated) are used

26 (e) Data from 2003 used due to lack of available data for 2004 and 2005

27 (f) Data for 1998 not available and values from 1999 are used  
28

1 **Table 2.2.** Illustrating the periods of operation, spectral bands, and products available from various different satellite sensors that have been used to retrieve aerosol  
 2 properties.  
 3

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 $\mu\text{m}$ )	$\tau_{\text{aer}}$ , $\alpha$	1-channel retrieval gives $\tau_{\lambda=0.63}$ over ocean (Husar et al., 1997; Ignatov and Stowe, 2002) 2-channel using 0.63 $\mu\text{m}$ and 0.86 $\mu\text{m}$ gives $\tau_{\lambda=0.55}$ and $\alpha$ over ocean assuming mono-modal aerosol size distribution (Mishchenko et al., 1999) 2-channel using 0.63 $\mu\text{m}$ & 0.86 $\mu\text{m}$ gives $\tau_{\lambda=0.55}$ and $\alpha$ over dark forests and lake surfaces (Soufflet et al., 1997) 2-channel using 0.64 $\mu\text{m}$ and 0.83 $\mu\text{m}$ gives $\tau_{\lambda=0.55}$ and $\alpha$ over ocean assuming a bimodal aerosol size distribution (Higurashi and Nakajima, 1999; Higurashi et al., 2000)
TOMS <sup>a</sup> (Total Ozone Mapping Spectrometer)	1979–present	0.33 $\mu\text{m}$ , 0.36 $\mu\text{m}$	Aerosol Index, $\tau_{\text{aer}}$	Aerosol index to $\tau_{\text{aer}}$ 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 Jan 2005–present	8 bands [0.44–0.91 $\mu\text{m}$ ]	$\tau_{\text{aer}}$ , $\alpha$ , DRE	Multiple view angles and polarization capabilities. 0.67 $\mu\text{m}$ and 0.86 $\mu\text{m}$ 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 $\mu\text{m}$ ] and 3.9 $\mu\text{m}$	$\tau_{\text{aer}}$ , $\alpha$	0.67 $\mu\text{m}$ and 0.86 $\mu\text{m}$ retrieval gives $\tau_{\lambda=0.50}$ and $\alpha$ over ocean. Bi-modal aerosol size distribution assumed (Nakajima and Higurashi, 1998; Higurashi et al., 2000).
MODIS (Moderate resolution Imaging Spectrometer)	2000–present	12 bands 0.41–2.1 $\mu\text{m}$	$\tau_{\text{aer}}$ , $\alpha$ , 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., 2005a).
MISR (Multi-angle Imaging Spectro- Radiometer)	2000–present	4 bands [0.47–0.86 $\mu\text{m}$ ]	$\tau_{\text{aer}}$ , $\alpha$	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) $\tau_{\text{aer}}$ against upwelling irradiance Loeb and Kato (2002)
GLAS Geoscience Laser Altimeter System	2003–present	Active lidar [0.53, 1.06 $\mu\text{m}$ ]	Aerosol vertical profile	Lidar footprint roughly 70m at 170 m intervals. 8-day repeat orbiting cycle (Spinhirne et al., 2005).

ATSR-2/AATSR (Along Track Scanning Radiometer)	1996–present	4-bands [0.56–1.65 $\mu\text{m}$ ]	$\tau_{\text{aer}}, \alpha$	Nadir and 52° forward viewing geometry. 40 aerosol climatological mixtures containing up to six aerosol species are used in the retrievals (Veefkind et al., 1998; Holzer-Popp et al. 2002).
SeaWiFS (Sea-Viewing Wide Field-of-View Sensor)	1997-present	0.765 and 0.865 $\mu\text{m}$ (ocean) 0.41 to 0.67 (land)	$\tau_{\text{aer}}, \alpha$	2-channel using 0.765 $\mu\text{m}$ and 0.856 $\mu\text{m}$ gives $\tau_{\lambda=0.856}$ and $\alpha$ over ocean. Bi-modal aerosol size distribution assumed (M. Wang et al., 2005). Retrievals over land and ocean using six visible channels from 0.41 to 0.67 $\mu\text{m}$ (von Hoyningen-Huene, 2003; Lee et al., 2004) also developed.

- 1 Notes:  
2 DRE = direct radiative effect and includes both natural and anthropogenic sources. The Angstrom exponent,  $\alpha$ , is the wavelength dependence of  $\tau_{\text{aer}}$  and is defined by  $\alpha = -$   
3  $\ln(\tau_{\text{aer}\lambda_1}/\tau_{\text{aer}\lambda_2})/\ln(\lambda_1/\lambda_2)$  where  $\lambda_1$ =wavelength 1 and  $\lambda_2$ =wavelength 2.  
4 (a) TOMS followed up by OMI instrument on EOS-Aura satellite, launched July 2004.  
5

1 **Table 2.3.** The Direct aerosol Radiative Effect (DRE) estimated from satellite remote sensing studies  
 2 (adapted and updated from Yu et al., 2006).  
 3

Reference	Instrument	Data Analysed	Brief Description	Clear Sky DRE (W m <sup>-2</sup> ) -ocean
Bellouin et al. (2005)	MODIS/ TOMS/ SSMI	2002	MODIS fine and total $\tau_{\text{aer}}$ 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 (2006)	MODIS	Aug 2001–Dec 2003	Best prescribed aerosol model fitted to MODIS data. $\tau_{\text{aer}}$ 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	$\tau_{\text{aer}}$ 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
Haywood et al. (1999)	ERBE	Jul 1987 – Dec 1988	DRE diagnosed from GCM-ERBE TOA irradiances	-6.7
Mean (standard deviation)				-5.4 (0.9)

4

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

No	Model	LOAD [mg SO <sub>4</sub> m <sup>-2</sup> ]	$\tau_{\text{aer}}$ (0.55 $\mu\text{m}$ )	$\tau_{\text{aer ant}}$ [%]	RF [W m <sup>-2</sup> ]	NRFM [W g <sup>-1</sup> ]	NRF [W m <sup>-2</sup> $\tau_{\text{aer}}^{-1}$ ]	Reference
<i>Published since IPCC, 2001</i>								
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	3.06			-0.32	-105		(Jacobson, 2001a)
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., 2004b)
L	UIO-GCM	2.28			-0.29	-127		(Kirkevåg and Iversen, 2002)
<i>AeroCom / identical emissions used for year 1750 and 2000</i>								
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	(Schulz et al. 2006)
Q	ECHAM5-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; Kirkevåg 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.** 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 using AeroCom emissions

No	Model	LOAD POM [mgPOM m <sup>-2</sup> ]	$\tau_{\text{aer}}$ POM	$\tau_{\text{aer}}$ POM ant	LOAD BC [mgm <sup>-2</sup> ]	RF BCPOM [W m <sup>-2</sup> ]	RF POM [W m <sup>-2</sup> ]	RF BC [W m <sup>-2</sup> ]	RF FFPOM [W m <sup>-2</sup> ]	RF FFBC [W m <sup>-2</sup> ]	RF BB [W m <sup>-2</sup> ]	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 <sup>a</sup>	0.18 <sup>a</sup>	-0.05 <sup>a</sup>	(Koch, 2001)
E	GISS	2.39			0.39	0.32	-0.18	0.50	-0.05 <sup>a</sup>	0.25 <sup>a</sup>	0.12 <sup>a</sup>	(Chuang and Seinfeld., 2002)
F	GISS	2.49			0.43	0.30	-0.23	0.53	-0.06 <sup>a</sup>	0.27 <sup>a</sup>	0.09 <sup>a</sup>	(Liao and Seinfeld, 2005)
G	SPRINT	2.67	0.029	82%	0.53	0.15	-0.27	0.42	-0.07 <sup>a</sup>	0.21 <sup>a</sup>	0.01 <sup>a</sup>	(Takemura et al., 2005)
H	GATORG	2.55			0.39	0.47	-0.06	0.55	-0.01 <sup>a</sup>	0.27 <sup>a</sup>	0.22 <sup>a</sup>	(Jacobson, 2001b)
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				(Kirkevåg and Iversen, 2002)
L	UMI	1.16	0.0060	53%	0.19	0.02	-0.23	0.25	-0.06 <sup>a</sup>	0.12 <sup>a</sup>	-0.01	(Liu and Penner, 2002)
M	UIO_CTM	1.12	0.0058	55%	0.19	0.02	-0.09 <sup>c</sup>	0.10 <sup>c</sup>	-0.04	0.11	-0.05	(Myhre et al., 2003)
N	LOA	1.41	0.0085	52%	0.25	0.14 <sup>b</sup>	-0.16 <sup>b</sup>	0.27 <sup>b</sup>	-0.04 <sup>a</sup>	0.13 <sup>a</sup>	0.05 <sup>a</sup>	(Reddy and Boucher, 2004)
O	LSCE	1.50	0.0079	46%	0.25	0.13	-0.17	0.30	-0.04 <sup>a</sup>	0.15 <sup>a</sup>	0.02 <sup>a</sup>	(Schulz et al., 2006)
P	ECHAM5-HAM	1.06 <sup>c</sup>	0.0028 <sup>c</sup>	55% <sup>c</sup>	0.18 <sup>c</sup>	0.34 <sup>b</sup>	-0.39 <sup>b</sup>	0.64 <sup>b</sup>	-0.05 <sup>c</sup>	0.16 <sup>c</sup>	0.23 <sup>a</sup>	(Stier et al., 2005)
Q	GISS	1.22	0.0060	51%	0.24	0.08	-0.14	0.22	-0.03 <sup>a</sup>	0.11 <sup>a</sup>	0.01 <sup>a</sup>	(Koch, 2001)
R	UIO_GCM	0.88	0.0046	59%	0.19	0.24	-0.06	0.36	-0.02 <sup>a</sup>	0.18 <sup>a</sup>	0.08 <sup>a</sup>	(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 <sup>d</sup>	-0.07	0.16	-0.02 <sup>a</sup>	0.08 <sup>a</sup>	0.03 <sup>a</sup>	(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	-0.16	0.29	-0.03	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=POMx0.25 ; FFBC=BCx0.5 ; BB=(BCPOM)-(FFPOM+FFBC)

(b) Models L,O,Q-T are used to provide a split in components : BCPOM=Total aerosol - SO<sub>4</sub> ; POM=BCPOMx(-1.15) ; BC=BCPOMx1.9

(c) ECHAM5-HAM fossil fuel carbonaceous forcing of -0.11 W m<sup>-2</sup>, POMant=POMx0.55; Bcant=BCx0.8

(d) ULAQ all sky values are taken to be 0.3 of their clear-sky values ; GISS absorption coefficient assumed to equal that of AeroCom models (7.4 m<sup>2</sup> g<sup>-1</sup>)

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

1 **Table 2.6.** Quantities related to estimates of the aerosol direct RF. Recent estimates of anthropogenic aerosol load (LOAD), anthropogenic aerosol optical depth  
 2 ( $\tau_{\text{aer}}$ ), its fraction of the present day total aerosol optical depth ( $\tau_{\text{aer ant}}$ ), cloud cover in aerosol model, total aerosol direct radiative forcing (RF) for clear sky and all  
 3 sky conditions, *surface forcing* and atmospheric all-sky forcing.

No	Model	LOAD	$\tau_{\text{aer}}$ (0.55 $\mu\text{m}$ )	$\tau_{\text{aer ant}}$ (0.55 $\mu\text{m}$ )	Cloud Cover	RF top clear sky	RF top all sky	Surface Forcing all sky	Atmospheric Forcing all sky	Reference
		[mg m <sup>-2</sup> ]		[%]	[%]	[W m <sup>-2</sup> ]	[W m <sup>-2</sup> ]	[W m <sup>-2</sup> ]	[W m <sup>-2</sup> ]	
<i>Published since IPCC, 2001</i>										
A	GISS	5.0			79%		-0.39 <sup>a</sup> +0.01 <sup>b</sup>	-1.98 <sup>a</sup> -2.42 <sup>b</sup>	1.59 <sup>a</sup> 2.43 <sup>b</sup>	(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	6.4*			62%	-0.89	-0.12	-2.5	2.38	(Jacobson, 2001a)
F	GISS	6.7	0.049				-0.23			(Hansen et al., 2005)
G	GISS	5.6	0.040				-0.63			(Koch, 2001)
<i>AeroCom / identical emissions used for year 1750 and 2000</i>										
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	1.14	(Reddy and Boucher, 2004)
K	LSCE	4.8	0.033	40%	62%	-0.94	-0.28	-0.93	0.66	(Schulz et al., 2006)
L	ECHAM5	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	-2.21	2.07	
Average H-P		3.8	0.030	26%	63%	-0.60	-0.18	-1.03	0.85	
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	0.17	
Average A-P		4.3	0.035	29%	64%	-0.63	-0.21	-1.21	1.26	
Stddev A-P		1.3	0.012	13%	7%	0.26	0.18	0.44	0.65	
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	2.43	

4 Notes:

5 (a) External mixture (b) Internal mixture

6 \*The load excludes that of mineral dust, some of which was considered anthropogenic in Jacobson (2001a).

1 **Table 2.7** Published model studies of the RF due to cloud albedo effect, in the context of liquid water clouds, with a listing of the relevant modelling details.  
2

Model	Model type	Aerosol species	Aerosol mixtures	Cloud types included	Microphysics	Radiative Forcing ( $Wm^{-2}$ )
Lohmann et al. (2000)	AGCM + sulphur 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) <b>-0.45 (albedo)</b> ----- -1.5 (total)
Jones et al. (2001)	AGCM + sulphur 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) <b>-1.34 (albedo)</b>
Williams et al. (2001b)	GCM with slab ocean + sulphur cycle (Hadley) ----- AGCM fixed SST	S, SS	E	stratiform and shallow Cu	Jones et al. (2001)	-1.69 (total) <b>-1.37 (albedo)</b> ----- -1.62 (total) <b>-1.43(albedo)</b>
Rotstayn and Penner (2001)  ----- Rotstayn and Liu (2003)	AGCM (CSIRO) fixed SST and sulphur loading  ----- Interactive sulphur cycle	S	-	warm and mixed phase	Rotstayn (1997); Rotstayn et al. (2000)  ----- Inclusion of dispersion	<b>-1.39 (albedo)</b>  ----- 12-35% decrease <b>-1.12 (albedo, mid value decreased)</b>
Ghan et al. (2001)	AGCM (PNL) + 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) <b>-0.85 (albedo)</b>
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 + mixed phase	Modified from Chuang and Penner (1995), no collision/coalescence	<b>-1.85 (albedo)</b>
Menon et al. (2002b)	GCM (GISS) + sulphur cycle	S,OC, SS	E	warm	Droplet number conc. and LWC, Del Genio et al. (1996), Sundqvist et al. (1989) warm and mixed phase,	-2.41 (total) <b>-1.55 (albedo)</b>

	fixed SST				improved vertical distribution of clouds (but only 9 layers). Global aerosol burdens poorly constrained	
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) <b>-1.35 (albedo)</b>
Suzuki et al. (2004)	AGCM (Japan) fixed SST	S, OC, BC, SS	E	stratiform	Berry(1967), Sundqvist(1978)	<b>0.54 (albedo)</b>
Quaas et al. (2004)	AGCM (LMDZ) + interactive sulphur cycle. Fixed SST	S	-	warm and mixed phase	Aerosol mass and cloud droplet number concentration, Boucher and Lohmann, (1995); Boucher et al. (1995)	<b>-1.3 (albedo)</b>
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, 2006)	<b>-0.77 (albedo)</b>
Kristjansson et al. (2005)	CCM3 (NCAR)+ sulphur 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 sulphur 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 ----- fit to POLDER data fit to MODIS data	<b>-0.9 (albedo)</b> ----- <b>-0.5 (albedo)*</b> <b>-0.3 (albedo) *</b>
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)* -0.29 (total (ECHAM4)*
Dufresne et al. (2005)	AGCM (LMDZ) + interactive sulphur cycle fixed SST	S	-	warm	Aerosol mass and cloud droplet number concentration, Boucher and Lohmann, (1995), fitted to POLDER data	<b>-0.22 (albedo) *</b>
Takemura et	AGCM	S, OC, BC, SS,	E (50% BC from fossil	warm	Activation based on Kohler theory and updraft velocity	-0.94 (total)

al. (2005)	(SPRINTARS) + slab ocean	D	fuel); I (for OC and BC)			<b>-0.52 (albedo)</b>
Chen and Penner (2005)	AGCM (UM) + fixed SST	S, SS, D, OC, BC	I	warm + mixed phase	Aerosol mass and cloud droplet number concentration (log-normal) CTRL (Abdul-Razzak and Ghan, 2002)  B_RL_up B_RL_med Updraft velocity (W_TKE) B_RL_low Chuang et al. (1997, Para_CP) Nenes and Seinfeld (2003; Para_NS)	<b>-1.30 (albedo-ctl)</b> <b>-0.75 (albedo-1)</b> <b>-0.86 (albedo-2)</b> <b>-1.07 (albedo-3)</b> <b>-1.10 (albedo-4)</b> <b>-1.29 (albedo-5)</b> <b>-1.79 (albedo-6)</b>
Ming et al. (2005b)	AGCM (GFDL) (fixed SST and sulphur loading)	S	-	warm	Rotstayn et al. (2000), Khainroutdinov and Kogan (2000) (aerosols offline)	-2.3 (total) <b>-1.4 (albedo)</b>
Penner et al. (2006a)	LMDZ Oslo CCSR	S, SS, D, OC, BC	E	warm + mixed phase	Aerosol mass and cloud droplet number concentration; Boucher and Lohmann, (1995), Chen and Penner(2005), Sundqvist (1978) (results from experiment 1)	<b>-0.65 (albedo LMDZ)</b> <b>-0.68 (albedo Oslo)</b> <b>-0.74 (albedo CCSR)</b>

1  
2  
3  
4  
5  
6

Notes:

In the "Aerosol species" column S refers to sulphate, SS refers to sea salt, D refers to mineral dust, BC refers to black carbon, OC refers to organic carbon, and N refers to nitrate

In the "Aerosol mixtures" column E refers to external mixtures and I refers to internal mixtures

Only the bold numbers in the last column are used to construct Figure 2.16.

\* 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<sub>2</sub> rise from anthropogenic land cover change, and RF due to land use change-induced CO<sub>2</sub> 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 7.2)

Main Source of Land Cover Data	Forest Area PNV 10 <sup>6</sup> km <sup>2</sup>	Forest Area circa 1700 10 <sup>6</sup> km <sup>2</sup>	Forest Area circa 1990 10 <sup>6</sup> km <sup>2</sup>	Contribution to CO <sub>2</sub> Rise 1850–2000 <sup>a</sup> (ppm)	CO <sub>2</sub> RF <sup>a</sup> (W m <sup>-2</sup> )	Albedo RF vs PNV (W m <sup>-2</sup> )	Albedo RF vs 1750 (W m <sup>-2</sup> )
Ramankutty and Foley (1999)	55.27	52.77 <sup>n</sup>	43.97 <sup>b</sup>	16 <sup>c</sup>	0.27	-0.24 <sup>d</sup> -0.29 - +0.02 <sup>e</sup> -0.2 <sup>f</sup>	-0.18 <sup>d</sup> -0.22 - +0.02 <sup>q</sup> -0.14 <sup>g</sup> -0.15 - -0.28 <sup>g,h</sup> -0.15 <sup>o</sup> -0.075 - -0.325 <sup>g,t</sup>
Klein Goldewijk (2001)	58.6	54.4	41.5	12 <sup>c</sup>	0.20	-0.66 - +0.1 <sup>e</sup>	-0.50 - +0.08 <sup>q</sup> -0.275 <sup>g,t</sup>
Houghton (1983 <sup>m</sup> , 2003)		62.15	50.53 <sup>i</sup>	35 <sup>c</sup> 26 <sup>s</sup>	0.57 0.44		
MODIS (Schaaf et al., 2002)						-0.09 <sup>f</sup>	-0.07 <sup>q</sup>
Wilson and Henderson-Sellers (1985)						-0.2 <sup>j</sup>	-0.15 <sup>q</sup>
SARB <sup>p</sup>						-0.29 <sup>e</sup>	-0.22 <sup>q</sup>
Matthews (1983)						-0.11 - -0.55 <sup>e</sup>	-0.08 - -0.41 <sup>q</sup>
						-0.12 <sup>e</sup>	-0.09 <sup>q</sup>
						-0.4 <sup>k</sup>	-0.3 <sup>q</sup>
						-0.08 <sup>l</sup>	-0.06 <sup>q</sup>

Notes:

(a) CO<sub>2</sub> RFs at 2000 relative to 1850, calculated from the land use change contribution to the total increase in CO<sub>2</sub> from 1850 to 2000 simulated with both land use and fossil fuel emissions by the carbon cycle models

(b) 1992 forest area

(c) Land use contribution CO<sub>2</sub> rise from Brovkin et al. (2004)

(d) Albedo RF from Betts et al. (2006). Land cover data combined from Ramankutty and Foley (1999), Klein Goldewijk (2001) and Wilson and Henderson-Sellers (1985)

(e) Albedo RF from Myhre and Myhre (2003). Range of estimate for each land cover dataset arises from use of different albedo values

(f) Albedo RF from model of Goosse et al. (2005) in Brovkin et al. (2006)

(g) Estimate relative to 1700

(h) Albedo RF from Matthews et al. (2003)

(i) 1980 forest area

(j) Albedo RF from Betts (2001)

(k) Albedo RF from Hansen et al. (1997)

(l) Albedo RF from Govindasamy et al. (2001a)

(m) Forest areas aggregated by Richards (1990)

(n) 1750 forest area reported as 51.85 10<sup>6</sup> km<sup>2</sup>

(o) Albedo RF from Hansen et al. (2005)

- 1 (p) <http://www-surf.larc.nasa.gov/surf/>
- 2 (q) RF relative to 1750 estimated here as 0.75 of RF relative to PNV following Betts et al. (2006) and Brovkin et al. (2006)
- 3 (r) Albedo RF from Myhre et al. (2005b)
- 4 (s) Land use contribution to CO<sub>2</sub> rise from Matthews et al. (2004). Estimate only available relative to 1850 not 1750.
- 5 (t) Albedo RF from Matthews et al. (2004)
- 6 (u) The available literature simulates CO<sub>2</sub> rises with and without land use relative to 1850
- 7

1 **Table 2.9.** Radiative forcing terms for contrail and cirrus effects caused by global subsonic aircraft  
 2 operations  
 3

Radiative forcing ( $\text{W m}^{-2}$ ) <sup>a</sup>	1992 IPCC <sup>b</sup>	2000 IPCC <sup>c</sup>	2000 <sup>d</sup>
CO <sub>2</sub> <sup>d</sup>	0.018	0.025	0.025
Persistent contrails	0.020	0.034	0.010 (0.006–0.015)
Aviation-induced cloudiness without persistent contrails	0–0.040	–	
Aviation-induced cloudiness (with persistent contrails)			0.030 (0.010–0.080)

4 Notes:

5 (a) Values for contrails are best estimates. Values in parentheses give the uncertainty range.

6 (b) Values from IPCC-1999 (IPCC, 1999)

7 (c) Values interpolated from 1992 and 2015 estimates in IPCC-1999 (Sausen et al., 2005)

8 (d) Sausen et al. (2005). Values are considered valid (within 10%) for 2005 because of slow growth in aviation fuel use  
 9 between 2000 and 2005.

10

1 **Table 2.10.** Comparison of the estimates of the increase in RF from the seventeenth century Maunder  
 2 Minimum (MM) to contemporary solar minima, documenting new understanding since the TAR.

Reference	Assumptions and Technique	RF from the Maunder Minimum to Contemporary Minima ( $\text{W m}^{-2}$ )	Comment on Current Understanding
Schatten and Orosz (1990)	Extrapolation of the 11-year irradiance cycle to the MM, using the sunspot record.	Approx. 0	Irradiance levels at cycle minima remain approximately constant.
Lean, Skumanich, and White (1992)	No spots, plage or network in Ca images assumed during MM.	0.26	Maximum irradiance increase from a non-magnetic sun, due to changes in known bright features on contemporary solar disk.
Lean, Skumanich, and White (1992)	No spots, plage or network and reduced basal emission in cell centres in Ca images to match reduced brightness in non-cycling stars, assumed to be MM analogs.	0.45	New assessment of stellar data (Hall and Lockwood, 2004) does not support original stellar brightness distribution, nor the use of the brightness reduction in the Baliunas and Jastrow (1990) "non-cycling" stars as MM analogs.
Hoyt and Schatten (1993)*	Convective restructuring implied by changes in sunspot umbra/penumbra ratios from MM to present: amplitude of increase from MM to present based on brightness of non-cycling stars, from Lean et al. (1992).	0.65	As above
Lean et al. (1995)	Reduced brightness of non-cycling stars, relative to those with active cycles, assumed typical of MM.	0.45	As above
Solanki and Fligge (1999)*	Combinations of above.	0.68	As above
Lean (2000)	Reduced brightness of non-cycling stars (revised solar-stellar calibration) assumed typical of MM.	0.38	As above
Foster (2004) Model	Non-magnetic sun estimates by removing bright features from MDI images assumed for MM.	0.28	Similar approach to removal of spots, plage and network by Lean et al. (1992).
Y Wang et al. (2005)*	Flux transport simulations of total magnetic flux evolution from MM to present.	0.1	Solar model suggests that modest accumulation of magnetic flux from one solar cycle to the next produces a modest increase in irradiance levels at solar cycle minima.
Dziembowski et al. (2001)	Helioseismic observations of solar interior oscillations suggest that the historical Sun cannot be any dimmer than current activity minima.	Approx. 0	

3 Notes:

4 The RF is the irradiance change divided by 4 (geometry) and multiplied by 0.7 (albedo). The solar activity cycle, which  
 5 was negligible during the Maunder Minimum and is of order  $1 \text{ W m}^{-2}$  (min-to-max) during recent cycles, is  
 6 superimposed on the irradiance changes at cycle minima. When smoothed over 20 years, this cycle increases the net RF  
 7 in the table by an additional  $0.09 \text{ W m}^{-2}$ . Reconstructions identified by \* extend only to 1713, the end of the Maunder  
 8 Minimum.  
 9

1 **Table 2.11.** Uncertainty assessment of forcing agents discussed in this chapter. Uncertainties are in approximate order of importance with first-order uncertainties  
 2 listed first.  
 3

	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	A	3	Low	Global trends since 1990; methane contribution to trend; Spectroscopy	Global trends prior to 1990; radiative transfer in climate models; CTM models of methane oxidation	Range based on uncertainties in methane contribution to trend and published RF estimates
Direct aerosol	A	2-3	Medium-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 effect (all aerosols)	B	3	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 and published results where models have been constrained by satellite data .
Surface albedo (land use)	A	2-3	Medium-Low	Some quantification of deforestation and desertification.	Separation of anthropogenic changes from natural;	Based on range of published estimates and published uncertainty analyses.
Surface albedo (black-carbon aerosol on snow)	B	3	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 on a few published model studies
Persistent linear 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
Solar irradiance	B	3	Low	Measurements over last 25 years; proxy indicators of solar activity	Relationship between proxy data and total solar irradiance; indirect ozone effects	Range from available reconstructions of solar irradiance and their qualitative

						assessment
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
Stratospheric water vapour from other	C	3	Very 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	Very Low	Process understood; spectroscopy; some regional information	Global injection poorly quantified	Not given
Aviation induced cirrus	C	3	Very Low	Cirrus radiative and microphysical properties; aviation emissions; contrail coverage in certain regions	transformation of contrails to cirrus; Aviation's effect on cirrus clouds;	Not given
Cosmic rays	C	3	Very 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
Other surface effects	C	3	Very 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 given

1  
2  
3  
4  
5  
6

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

**Table 2.12.** Global mean radiative forcings since 1750 and comparison with earlier assessments

	Global mean radiative forcing (W m <sup>-2</sup> ) ±90% value uncertainty range			Summary comments on changes since the TAR
	SAR (1750-1993)	TAR (1750-1998)	AR4 (1750-2005)	
<b>Combined Anthropogenic RF</b>	Not evaluated	Not evaluated	1.6 [-1.0, +0.8]	<b>Newly evaluated. Probability density function estimate</b>
<b>Long Lived Greenhouse gases {Comprising CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, and halocarbons}</b>	+2.45 [15%] {CO <sub>2</sub> (1.56); CH <sub>4</sub> (0.47); N <sub>2</sub> O (0.14); Halocarbons (0.28)}	+2.43 [10%] {CO <sub>2</sub> (1.46); CH <sub>4</sub> (0.48); N <sub>2</sub> O (0.15); Halocarbons (0.34) <sup>1</sup> }	+2.63 [± 0.26] {CO <sub>2</sub> : 1.66 [± 0.17]); CH <sub>4</sub> (0.48 [± 0.05]); N <sub>2</sub> O (0.16 [± 0.02]); Halocarbons (0.34 [± 0.03])}	<b>Total increase in RF, due to upward trends, particularly in CO<sub>2</sub>. Halocarbon RF trend is positive<sup>1</sup></b>
Stratospheric O <sub>3</sub>	-0.1 [2X]	-0.15 [67%]	-0.05 [± 0.10]	<b>Re-evaluated to be weaker</b>
Tropospheric O <sub>3</sub>	+0.40 [50%]	+0.35 [43%]	+0.35 [-0.1, +0.3]	<b>Best estimate unchanged. However, a larger RF could be possible</b>
Stratospheric water vapour from CH <sub>4</sub>	Not evaluated	+0.01 to +0.03	+0.07 [± 0.05]	<b>Re-evaluated higher</b>
<b>Total direct aerosol</b>	Not evaluated	Not evaluated	-0.50 [± 0.40]	<b>Newly evaluated</b>
Direct sulphate aerosol	-0.40 [2X]	-0.40 [2X]	-0.40 [± 0.20]	Better constrained
Direct FF aerosol (OC)	Not evaluated	-0.10 [3X]	-0.05 [± 0.05]	Re-evaluated to be weaker
Direct FF aerosol (BC)	+0.10 [3X]	+0.20 [2X]	+0.20 [± 0.15]	Similar best estimate to the TAR. Response affected by semi-direct effects
Direct biomass burning aerosol	-0.20 [3X]	-0.20 [3X]	+0.05 [± 0.13]	Re-evaluated and sign changed. Response affected by semi-direct effects
Direct nitrate aerosol	Not evaluated	Not evaluated	-0.10 [± 0.10]	Newly evaluated
Direct mineral dust aerosol	Not evaluated	-0.60 to +0.40	-0.10 [± 0.20]	Re-evaluated to have a smaller anthropogenic fraction
<b>Cloud albedo effect</b>	<b>0 to -1.5 (sulphate only)</b>	<b>0.0 to -2.0 (all aerosols)</b>	<b>-0.70 [-1.1, +0.4] (all aerosols)</b>	<b>Best estimate now given</b>
Surface albedo (land-use)	Not evaluated	-0.20 [100%]	-0.20 [± 0.20]	<b>Additional studies</b>
Surface albedo (black-carbon aerosol on snow)	Not evaluated	Not evaluated	+0.10 [±0.10]	<b>Newly evaluated</b>
Persistent linear contrails	Not evaluated	0.02 [3.5 X]	0.01 [-0.007, +0.02]	<b>Re-evaluated to be smaller</b>
Solar irradiance	+0.30 [67%]	+0.30 [67%]	+0.12 [-0.06, +0.18]	<b>Re-evaluated to be less than half</b>

Notes:

Bold rows appear on Figure 2.20.

The first row shows the combined anthropogenic RF from the probability density function in Panel B of Figure 2.20.

The sum of the individual RFs and their estimated errors are not quite the same as the numbers presented in this row due to the statistical construction of the probability density function.

1 For the AR4 column 90% value uncertainties appear in brackets: when adding these numbers to the best estimate the 5–  
2 95% confidence range is obtained. When two numbers are quoted for the value uncertainty the distribution is non-  
3 normal. Uncertainties in the SAR and the TAR had a similar basis, but their evaluation was more subjective. [2x], [3x]  
4 etc. refers to a factor of two uncertainty and a log-normal distribution of RF estimates.  
5 <sup>1</sup> The TAR RF for halocarbons and hence the total LLGHG RF was incorrectly evaluated some 0.01 W m<sup>-2</sup> too high.  
6 The actual trends in these RFs are therefore more positive than suggested by numbers in this table (Table 2.1 shows  
7 updated trends).  
8

1 **Table 2.13.** Emission based RFs for emitted components with radiative effects other than through changes in their atmospheric abundance. Minor effects where the  
 2 estimated RF is less than  $0.01 \text{ W m}^{-2}$  are not included. Effects on sulphate aerosols are not included since  $\text{SO}_2$  emission is the only significant factor effecting  
 3 sulphate aerosols. Method of calculation and uncertainty ranges are given in the footnotes.  
 4

		Atmospheric or surface change directly causing radiative forcing													
		CO <sub>2</sub>	CH <sub>4</sub>	CFC/ HCFC	N <sub>2</sub> O	HFC/ PFC /SF <sub>6</sub>	BC- direct	BC snow albedo	OC	O <sub>3</sub> (T)	O <sub>3</sub> (S)	H <sub>2</sub> O(S)	Nitrate aerosols	Indirect cloud albedo effect	
<b>Component emitted</b>	CO <sub>2</sub>	1.56 <sup>a</sup>													
	CH <sub>4</sub>	0.016 <sup>a</sup>	0.57 <sup>c</sup>							0.2 <sup>c</sup>		0.07 <sup>d</sup>			
	CFC/HCFC/halons			0.32 <sup>h</sup>									-0.065 <sup>b</sup>		
	N <sub>2</sub> O				0.15 <sup>h</sup>								-0.035 <sup>b</sup>		
	HFC/PFC/ SF <sub>6</sub>					0.017 <sup>h</sup>									
	CO/VOC	0.06 <sup>a</sup>	0.08 <sup>c</sup>								0.13 <sup>c</sup>				
	NO <sub>x</sub>		-							0.06 <sup>c</sup>			-0.10 <sup>e</sup>	X <sup>i</sup>	
			0.17 <sup>c</sup>												
	BC						0.37 <sup>f</sup>	0.1 <sup>g</sup>							X <sup>i</sup>
	OC								-0.2 <sup>f</sup>						X <sup>i</sup>
														X <sup>i</sup>	
	SO <sub>2</sub>													X <sup>i</sup>	

- 5 Notes:  
 6 (a) RF of the observed CO<sub>2</sub> change (Table 2.12), and then subtracted the contributions from CH<sub>4</sub>, CO and VOC emissions from fossil sources. Historic emissions of CH<sub>4</sub>, CO and  
 7 VOCs from EDGAR-HYDE (Van Aardenne et al. (2001)), CO<sub>2</sub> contribution from these sources calculated by with CO<sub>2</sub> model described by Joos et al. (1996).  
 8 (b) 80% of RF from observed ozone depletion in the stratosphere (Table 2.12) is attributed to CFCs/HFCs, remaining 20% to N<sub>2</sub>O (Based on Nevison et al. (1999) and WMO (2003))  
 9 (c) RF of the observed CH<sub>4</sub> change (Table 2.12), and then subtracted the contributions through lifetime changes through OH changes caused by emissions of NO<sub>x</sub>, CO and VOC.  
 10 The effects of NO<sub>x</sub>, CO and VOCs are from Shindell et al. (2005). There are significant uncertainties related to these relations. Following Shindell et al. (2005) the uncertainty  
 11 estimate is taken to be  $\pm 20\%$  for methane emissions, and  $\pm 50\%$  for CO, VOC and NO<sub>x</sub> emissions.  
 12 (d) All the radiative forcing from changes in stratospheric water vapour is attributed to methane emissions (Section 2.3.7 and Table 2.12)  
 13 (e) RF cf. Table 2.12, uncertainty  $\pm 0.10 \text{ W m}^{-2}$   
 14 (f) Mean of all studies in Table 2.5, includes FF, biofuel and biomass burning, Uncertainty (90% confidence ranges)  $0.50 \text{ W m}^{-2}$  (BC) and  $0.30 \text{ W m}^{-2}$  (OC) based on range of reported  
 15 values in Table 2.5.  
 16 (g) RF cf. Table 2.12, uncertainty  $\pm 0.10 \text{ W m}^{-2}$   
 17 (h) RF calculated based on observed concentration change, cf. Table 2.12 and Section 2.3  
 18 (i) Uncertainty too large to apportion the indirect cloud albedo effect to each aerosol type (Hansen et al., 2005)

1 **Table 2.14.** Lifetimes, radiative efficiencies, and direct (except for methane) global warming potentials (GWP) relative to carbon dioxide. For ozone depleting  
 2 substances and their replacements data are taken from WMO (2003) unless otherwise indicated.  
 3

Industrial Designation or Common Name	Chemical Formula	Other Name	Lifetime (years)	Radiative Efficiency ( $\text{W m}^{-2} \text{ ppb}^{-1}$ )	Global Warming Potential for Given Time Horizon (years)			
					SAR (100)	20	100	500
Carbon dioxide	CO <sub>2</sub>		See below <sup>a</sup>	<sup>b</sup> 1.4x10 <sup>-5</sup>	1	1	1	1
Methane <sup>c</sup>	CH <sub>4</sub>		12 <sup>c</sup>	3.7x10 <sup>-4</sup>	21	72	25	7.6
Nitrous oxide	N <sub>2</sub> O		114 <sup>c</sup>	3.03x10 <sup>-3</sup>	310	289	298	153
<i>Substances controlled by the Montreal Protocol</i>								
CFC-11	CCl <sub>3</sub> F	Trichlorofluoromethane	45	0.25	3800	6730	4750	1620
CFC-12	CCl <sub>2</sub> F <sub>2</sub>	Dichlorodifluoromethane	100	0.32	8100	11000	10900	5200
CFC-13	CClF <sub>3</sub>	Chlorotrifluoromethane	640	0.25		10800	14400	16400
CFC-113	CCl <sub>2</sub> FCClF <sub>2</sub>	1,1,2-Trichlorotrifluoroethane	85	0.3	4800	6540	6130	2700
CFC-114	CClF <sub>2</sub> CClF <sub>2</sub>	Dichlorotetrafluoroethane	300	0.31		8040	10000	8730
CFC-115	CClF <sub>2</sub> CF <sub>3</sub>	Monochloropentafluoroethane	1700	0.18		5310	7370	9990
Halon-1301	CBrF <sub>3</sub>	Bromotrifluoromethane	65	0.32	5400	8480	7140	2760
Halon-1211	CBrClF <sub>2</sub>	Bromochlorodifluoromethane	16	0.3		4750	1890	575
Halon-2402	CBrF <sub>2</sub> CBrF <sub>2</sub>	1,2-Dibromotetrafluoroethane	20	0.33		3680	1640	503
Carbon tetrachloride	CCl <sub>4</sub>	(Halon-104)	26	0.13	1400	2700	1400	435
Methyl bromide	CH <sub>3</sub> Br	(Halon-1001)	0.7	0.01		17	5	1
Methyl chloroform	CH <sub>3</sub> CCl <sub>3</sub>	1,1,1-Trichloroethane	5	0.06		506	146	45
HCFC-22	CHClF <sub>2</sub>	Chlorodifluoromethane	12	0.2	1500	5160	1810	549
HCFC-123	CHCl <sub>2</sub> CF <sub>3</sub>	Dichlorotrifluoroethane	1.3	0.14	90	273	77	24
HCFC-124	CHClF <sub>2</sub> CF <sub>3</sub>	Chlorotetrafluoroethane	5.8	0.22	470	2070	609	185
HCFC-141b	CH <sub>3</sub> CCl <sub>2</sub> F	Dichlorofluoroethane	9.3	0.14		2250	725	220
HCFC-142b	CH <sub>3</sub> CClF <sub>2</sub>	Chlorodifluoroethane	17.9	0.2	1800	5490	2310	705
HCFC-225ca	CHCl <sub>2</sub> CF <sub>2</sub> CF <sub>3</sub>	Dichloropentafluoropropane	1.9	0.2		429	122	37
HCFC-225cb	CHClF <sub>2</sub> CF <sub>2</sub> CF <sub>3</sub>	Dichloropentafluoropropane	5.8	0.32		2030	595	181
<i>Hydrofluorocarbons</i>								
HFC-23	CHF <sub>3</sub>	Trifluoromethane	270	0.19	11700	12000	14800	12200
HFC-32	CH <sub>2</sub> F <sub>2</sub>	Difluoromethane	4.9	0.11	650	2330	675	205
HFC-125	CHF <sub>2</sub> CF <sub>3</sub>	Pentafluoroethane	29	0.23	2800	6350	3500	1100
HFC-134a	CH <sub>2</sub> FCF <sub>3</sub>	1,1,1,2-Tetrafluoroethane	14	0.16	1300	3830	1430	435
HFC-143a	CH <sub>3</sub> CF <sub>3</sub>	1,1,1-Trifluoroethane	52	0.13	3800	5890	4470	1590
HFC-152a	CH <sub>3</sub> CHF <sub>2</sub>	1,1-Difluoroethane	1.4	0.09	140	437	124	38

HFC-227ea	CF <sub>3</sub> CHFCF <sub>3</sub>	1,1,1,2,3,3,3-Heptafluoropropane	34.2	0.26	2900	5310	3220	1040
HFC-236fa	CF <sub>3</sub> CH <sub>2</sub> CF <sub>3</sub>	1,1,1,3,3,3-Hexafluoropropane	240	0.28	6300	8100	9810	7660
HFC-245fa	CHF <sub>2</sub> CH <sub>2</sub> CF <sub>3</sub>	1,1,1,3,3-Pentafluoropropane	7.6	0.28		3380	1030	314
HFC-365mfc	CH <sub>3</sub> CF <sub>2</sub> CH <sub>2</sub> CF <sub>3</sub>	1,1,1,3,3-Pentafluorobutane	8.6	0.21		2520	794	241
HFC-43-10mee	CF <sub>3</sub> CHFCHFCF <sub>2</sub> CF <sub>3</sub>	1,1,1,2,2,3,4,5,5,5-Decafluoropentane	15.9	0.4	1300	4140	1640	500
<i>Perfluorinated compounds</i>								
	SF <sub>6</sub>	Sulphur hexafluoride	3200	0.52	23900	16300	22800	32600
	NF <sub>3</sub>	Nitrogen trifluoride	740	0.21 <sup>f</sup>		12300	17200	20700
PFC-14	CF <sub>4</sub>	Carbon tetrafluoride	50000	0.10 <sup>e</sup>	6500	5210	7390	11200
PFC-116	C <sub>2</sub> F <sub>6</sub>	Perfluoroethane	10000	0.26	9200	8630	12200	18200
PFC-218	C <sub>3</sub> F <sub>8</sub>	Perfluoropropane	2600	0.26	7000	6310	8830	12500
PFC-318	c-C <sub>4</sub> F <sub>8</sub>	Perfluorocyclobutane	3200	0.32	8700	7310	10300	14700
PFC-3-1-10	C <sub>4</sub> F <sub>10</sub>	Perfluorobutane	2600	0.33	7000	6330	8860	12500
PFC-4-1-12	C <sub>5</sub> F <sub>12</sub>	Perfluoropentane	4100	0.41		6510	9160	13300
PFC-5-1-14	C <sub>6</sub> F <sub>14</sub>	Perfluorohexane	3200	0.49	7400	6600	9300	13300
PFC-9-1-18	C <sub>10</sub> F <sub>18</sub>	Perfluorodecalin	>1000 <sup>d</sup>	0.56		>5500	>7500 <sup>d</sup>	>9500
	SF <sub>5</sub> CF <sub>3</sub>	Trifluoromethyl sulphur pentafluoride	800	0.57		13200	17700	21200
<i>Fluorinated ethers</i>								
HFE-125	CF <sub>3</sub> OCHF <sub>2</sub>		136	0.44		13800	14900	8490
HFE-134	CHF <sub>2</sub> OCHF <sub>2</sub>		26	0.45		12200	6320	1960
HFE-143a	CH <sub>3</sub> OCF <sub>3</sub>		4.3	0.27		2630	756	230
HCFE-235da	CF <sub>3</sub> CHClOCHF <sub>2</sub>		2.6	0.38		1230	350	106
HFE-245cb2	CF <sub>3</sub> CF <sub>2</sub> OCH <sub>3</sub>		5.1	0.32		2440	708	215
HFE-245fa2	CF <sub>3</sub> CH <sub>2</sub> OCHF <sub>2</sub>		4.9	0.31		2280	659	200
HFE-254cb2	CHF <sub>2</sub> CF <sub>2</sub> OCH <sub>3</sub>		2.6	0.28		1260	359	109
HFE-347mcc3	CF <sub>3</sub> CF <sub>2</sub> CF <sub>2</sub> OCH <sub>3</sub>		5.2	0.34		1980	575	175
HFE-347pcf2	CF <sub>3</sub> CH <sub>2</sub> OCF <sub>2</sub> CHF <sub>2</sub>		7.1	0.25		1900	580	175
HFE-356pcf3	CHF <sub>2</sub> CF <sub>2</sub> CH <sub>2</sub> OCHF <sub>2</sub>		3.6	0.39		1760	502	153
HFE-449sl	C <sub>4</sub> F <sub>9</sub> OCH <sub>3</sub>	(HFE-7100)	3.8	0.31		1040	297	90
HFE-569sf2	C <sub>4</sub> F <sub>9</sub> OC <sub>2</sub> H <sub>5</sub>	(HFE-7200)	0.77	0.3		207	59	18
HFE-43-10pccc124	CHF <sub>2</sub> OCF <sub>2</sub> OC <sub>2</sub> F <sub>4</sub> OCHF <sub>2</sub>	(H-Galden 1040x)	6.3	1.37		6320	1870	569
HFE-236ca12	CHF <sub>2</sub> OCF <sub>2</sub> OCHF <sub>2</sub>	(HG-10)	12.1	0.66		8000	2800	860
HFE-338pcc13	CHF <sub>2</sub> OCF <sub>2</sub> CF <sub>2</sub> OCHF <sub>2</sub>	(HG-01)	6.2	0.87		5100	1500	460
<i>Perfluoropoly ethers</i>								
PFPME <sup>g</sup>	CF <sub>3</sub> OCF(CF <sub>3</sub> )CF <sub>2</sub> OCF <sub>2</sub> OCF <sub>3</sub>		800	0.65		7620	10300	12400

Hydrocarbons and other compounds - Direct Effects								
Dimethylether	CH <sub>3</sub> OCH <sub>3</sub>		0.015	0.02		1	1	<<1
Methylenechloride	CH <sub>2</sub> Cl <sub>2</sub>	Dichloromethane	0.38	0.03		31	8.7	2.7
Methyl chloride	CH <sub>3</sub> Cl	Chloromethane	1.0	0.01		45	13	4

1 Notes:

2 (a) The CO<sub>2</sub> response function used in this report is based on the revised version of the Bern Carbon cycle model used in Chapter 10 of this report using a background CO<sub>2</sub>  
3 concentrations 378 ppm. The decay of a pulse of CO<sub>2</sub> with time  $t$  is given by

4  $a_0 + \sum_{i=1}^3 a_i \cdot e^{-t/\tau_i}$ , where  $a_0=0.217$ ,  $a_1=0.259$ ,  $a_2=0.338$ ,  $a_3=0.186$ ,  $\tau_1=172.9$  years,  $\tau_2=18.51$  years, and  $\tau_3=1.186$  years.

5 (b) The radiative efficiency of CO<sub>2</sub> is calculated using the IPCC (1990) simplified expression as revised in the TAR, with an updated background concentration value of 378 ppm  
6 and a perturbation of +1 ppb (see Section 2.10.2).

7 (c) The perturbation lifetime for methane is 12 years as in the TAR (see also Chapter 7, Section 7.4). The GWP for methane includes indirect effects from enhancements of ozone  
8 and stratospheric water vapour (see Section 2.10.3.1)

9 (d) Shine et al. (2005c), updated by the revised AGWP for CO<sub>2</sub>. The assumed lifetime of 1000 years is a lower limit.

10 (e) Hurley et al. (2005)

11 (f) Robson et al. (2006)

12 (g) Young et al. (2006)

13

14

1 **Table 2.15** Indirect GWPs (100) for 10 NMVOCs from Collins et al. (2002) and for NO<sub>x</sub> emissions (on N-  
 2 basis) from Derwent et al. (2001), Wild et al. (2001), Berntsen et al. (2005), and Stevenson et al. (2004).  
 3

Organic compound/Study	GWP <sup>CH4</sup>	GWP <sup>O3</sup>	GWP
Ethane (C <sub>2</sub> H <sub>6</sub> )	2.9	2.6	5.5
Propane (C <sub>3</sub> H <sub>8</sub> )	2.7	0.6	3.3
Butane (C <sub>4</sub> H <sub>10</sub> )	2.3	1.7	4.0
Ethylene (C <sub>2</sub> H <sub>4</sub> )	1.5	2.2	3.7
Propylene (C <sub>3</sub> H <sub>6</sub> )	-2.0	3.8	1.8
Toluene (C <sub>7</sub> H <sub>8</sub> )	0.2	2.5	2.7
Isoprene (C <sub>5</sub> H <sub>8</sub> )	1.1	1.6	2.7
Methanol (CH <sub>3</sub> OH)	1.6	1.2	2.8
Acetaldehyde (CH <sub>3</sub> CHO)	-0.4	1.7	1.3
Acetone (CH <sub>3</sub> COCH <sub>3</sub> )	0.3	0.2	0.5
Derwent et al. NH surface NO <sub>x</sub> <sup>b,c</sup>	-24	11	-12
Derwent et al. SH surface NO <sub>x</sub> <sup>b,c</sup>	-64	33	-31
Wild et al., industrial NO <sub>x</sub>	-44	32	-12
Berntsen et al., surface NO <sub>x</sub> Asia	-31 to -42 <sup>a</sup>	55 to 70 <sup>a</sup>	25 to 29 <sup>a</sup>
Berntsen et al., surface NO <sub>x</sub> Europe	-8.6 to -11 <sup>a</sup>	8.1 to 12.7	-2.7 to +4.1 <sup>a</sup>
Derwent et al., Aircraft NO <sub>x</sub> <sup>b,c</sup>	-145	246	100
Wild et al., Aircraft NO <sub>x</sub>	-210	340	130
Stevenson et al. Aircraft NO <sub>x</sub>	-159	155	-3

4 Notes:

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

6 (b) Corrected values as described in Stevenson et al. (2004).

7 (c) For January pulse emissions.  
 8