

## Chapter 7: Couplings Between Changes in the Climate System and Biogeochemistry

**Coordinating Lead Authors:** Guy Brasseur, Kenneth Denman

**Lead Authors:** Amnat Chidthaisong, Philippe Ciais, Peter Cox, Robert Dickinson, Didier Hauglustaine, Christoph Heinze, Elisabeth Holland, Daniel Jacob, Ulrike Lohmann, Srikanthan Ramachandran, Pedro Leite da Silva Diaz, Steven Wofsy, Xiaoye Zhang

**Contributing Authors:** David Archer, V. Arora, John Austin, D. Baker, Joe Berry, Gordon Bonan, Philippe Bousquet, Deborah Clark, V. Eyring, Johann Feichter, Pierre Friedlingstein, Inez Fung, Sandro Fuzzi, Sunling Gong, Alex Guenther, Ann Henderson-Sellers, Andy Jones, Bernd Kärcher, Mikio Kawamiya, Yadvinder Malhi, K. Masarie, Surabi Menon, J. Miller, P. Peylin, A. Pitman, Johannes Quaas, P. Rayner, Ulf Riebesell, C. Rödenbeck, Leon Rotstayn, Nigel Roulet, Chris Sabine, Martin Schultz, Michael Schulz, Will Steffen, Steve Schwartz, J. Lee-Taylor, Yuhong Tian, Oliver Wild, Liming Zhou.

**Review Editors:** Kansri Boonpragob, Martin Heimann, Mario Molina

**Date of Draft:** 12 August 2005

**Notes:** This is the TSU compiled version

### Table of Contents

Executive Summary .....	3
7.1 Introduction.....	6
7.1.1 <i>Terrestrial Ecosystems and the Climate System</i> .....	6
7.1.2 <i>The Carbon Cycle and the Climate System</i> .....	7
7.1.3 <i>Reactive Gases and the Climate System</i> .....	8
7.1.4 <i>Aerosol Particles and the Climate System</i> .....	9
7.2 The Changing Land Surface and Climate.....	9
7.2.1 <i>Introduction to Land Climate Response from Human Activities</i> .....	9
7.2.2 <i>How Does Land Surface Mediate Climate Change?</i> .....	9
7.2.3 <i>What Has Been Learned About Land Model Structures Since the TAR?</i> .....	12
7.2.4 <i>How Can We Quantitatively Relate Land Climate Changes to Atmospheric Inputs and Structural Change?</i> .....	13
7.2.5 <i>Functioning of System Models - Improving Their Basis for Quantitative Evaluation of Impacts of Land Changes</i> .....	16
7.3 The Carbon Cycle and the Climate System.....	17
7.3.1 <i>Contemporary Carbon Budget</i> .....	17
7.3.2 <i>Carbon Cycle Processes</i> .....	19
Box 7.1: Marine Carbon Chemistry and Ocean Acidification .....	23
7.3.3 <i>Interannual and Regional Variation in Carbon Fluxes Inferred from Observations</i> .....	32
7.3.4 <i>Coupled Climate-Carbon Cycle Projections</i> .....	38
Question 7.1: Are the Increases in Atmospheric Carbon Dioxide and Other Greenhouse Gases During the Industrial Era Caused by Human Activities? .....	44
7.4 Reactive Gases and the Climate System.....	45
7.4.1 <i>Methane (CH<sub>4</sub>)</i> .....	45
7.4.2 <i>Nitrogen Compounds</i> .....	50
7.4.3 <i>Molecular Hydrogen</i> .....	52
7.4.4 <i>Global Tropospheric Ozone</i> .....	53
7.4.5 <i>The Hydroxyl Radical (OH)</i> .....	57
7.4.6 <i>Stratospheric Ozone and Climate</i> .....	61
7.5 Aerosol Particles and the Climate System.....	62
7.5.1 <i>What Major Climatic Factors Control Natural Aerosol Emissions and Burdens?</i> .....	63
7.5.2 <i>How Do Aerosols (Indirect Effects) Modify Clouds and Precipitation?</i> .....	65
7.5.3 <i>How Are Changes in Aerosols/Clouds Affecting Processes at the Earth's Surface?</i> .....	70
7.5.4 <i>How Are Aerosols Affecting Circulation Patterns?</i> .....	72
7.6 Concluding Remarks .....	74

1	Box 7.2: Effects of Climate Change on Air Quality .....	74
2	References .....	77
3	Tables.....	118
4		

## Executive Summary

Biogeochemical cycles interact with the climate system over a variety of temporal and spatial scales. Nonlinear interactions involving physical, chemical and biological processes within or between the different compartments of the Earth system could amplify (positive feedbacks) or attenuate (negative feedbacks) the disturbances produced by human activities. The response of the climate system to anthropogenic perturbations is therefore expected to be more complex and more chaotic than a simple cause-effect relationship; it involves reciprocal interactions with the land surface, the carbon cycle, reactive gases and aerosol particles.

### *The Land Surface and Climate*

- Changes in temperature, moisture, and solar irradiance as well as atmospheric composition will affect the distribution and functioning of terrestrial ecosystems. The ecosystem response, in turn, will alter the momentum, energy, and water balance of the atmosphere, as well as the cycling of carbon, methane, dust and other atmospheric trace constituents.
- Changes in land vegetation, soils, and water cycle resulting from human activities can affect regional climate through shifts in cloudiness, surface temperatures, and radiation.
- Land use changes have contributed ~35% of the buildup of atmospheric CO<sub>2</sub> since preindustrial times – indicating that future scenarios and model projections must account for land use change.
- Many modelling studies demonstrate that changing land cover can have local and regional climate impacts that are comparable in magnitude with temperature and precipitation changes observed over the last several decades as reported in Chapter 3.
- New developments in dynamic vegetation schemes and coupled climate-carbon models have shown that the physiological forcing of stomatal closure can contribute 20% to the rainfall reduction in the Amazon associated with rising atmospheric CO<sub>2</sub> levels.

### *The Carbon Cycle and Climate*

- The average increase in atmospheric CO<sub>2</sub>, determined from the global CMDL network, was 1.88 ppm yr<sup>-1</sup>, or 3.99 Pg-C yr<sup>-1</sup> for 2000–2004.
- The land was a large source of CO<sub>2</sub> to the atmosphere in 1988 because of fires, and appeared to be a smaller sink for 1998–2003 because of the severe droughts.
- The airborne fraction of fossil fuel CO<sub>2</sub> appears to have increased in the last decade, and the partitioning of carbon sink between ocean and land appears to have shifted. However, it is premature to conclude if this is indicative of a long-term trend.
- New estimates, based on estimates of deforested areas from satellite images, yield a CO<sub>2</sub> source from land use modification of ~0.95±0.4 Pg-C yr<sup>-1</sup> for the decade of the 1990's. The satellite-based estimate represents approximately 50% of the previous estimates based on FAO statistics. This reduction of tropical land source implies a concomitant reduction in the tropical land sink to balance the global carbon budget.
- Interannual variability (~2–4 years) in atmospheric CO<sub>2</sub> concentrations results primarily from changes in net land uptake, not ocean uptake (from inverse analyses).
- Inverse analyses of observations show ocean uptake of CO<sub>2</sub> greater in Northern than Southern extratropics, while the tropics are outgassing (0–1.5 Pg-C yr<sup>-1</sup>)
- There is a net terrestrial sink of CO<sub>2</sub> in the Northern Hemisphere, with NH fluxes better constrained than those in the tropics.
- Interannual variability in global terrestrial CO<sub>2</sub> uptake is greater than ocean uptake; variability in terrestrial uptake is greater in the tropics than in temperate or boreal regions.
- Increasing CO<sub>2</sub> concentrations in the ocean are lowering the pH (increasing the acidity) with expected but poorly known consequences for marine ecosystems and sediments.
- Decreasing surface ocean buffer capacity for CO<sub>2</sub>, due to lowering pH and rising surface temperatures, reduces the rate at which the ocean can take up excess atmospheric CO<sub>2</sub>.
- The recent model intercomparison project (C<sup>4</sup>MIP) shows that, when the carbon cycle is coupled to a climate model, the atmospheric CO<sub>2</sub> buildup (scenario A2) is enhanced by ~20–180 ppm in year 2100 and temperature by 0–1.5°C relative to model predictions for which this coupling is ignored. Even

1 though all models exhibit a positive carbon-climate feedback, the magnitude of this feedback varies  
2 considerably between models.

- 3 • Differences in ocean uptake between models are due to differences in the removal rate of CO<sub>2</sub> from the  
4 surface ocean to greater depths due both to physical and biogeochemical processes.
- 5 • Uncertainties in terrestrial uptake and release of CO<sub>2</sub> are due to the magnitude of the fertilization effect,  
6 the land use legacy, limits on the growth and size of the terrestrial biosphere, and the turnover time of  
7 soil carbon.
- 8 • Terrestrial carbon storage depends on both temperature and moisture. Changes in temperature and  
9 moisture tend to co-vary, albeit with signs and magnitudes that depend on region and time scale.
- 10 • Overall, climate change will increase the fraction of anthropogenic CO<sub>2</sub> that remains in the atmosphere.  
11 Increasing CO<sub>2</sub> will increase the uptake by both the land and the ocean; climate change will cause  
12 reduced uptake by both the land and the ocean.

### 13 *Reactive Gases and Climate*

- 14 • The atmospheric burden of methane has not increased significantly during the period 2000–2005.  
15 Emissions of methane from energy use and from northern wetlands appear to have decreased, while  
16 emissions from lower latitudes have probably been increasing. This offset may explain the temporary  
17 slow down of the methane growth rate.
- 18 • Climate change could affect substantially the sources of methane, particularly those associated with  
19 wetlands and rice agriculture. Emissions are expected to increase in warmer and wetter climate and to  
20 decrease in warmer and dryer climate. The growing use of hydrogen-based energy should not affect  
21 stratospheric temperature and ozone in a substantial way. However, the use of hydrogen-powered  
22 vehicles should lead to a significant decrease in NO<sub>x</sub> emissions, and therefore in the concentration of  
23 tropospheric ozone and OH in urban areas.
- 24 • New model estimates of the global tropospheric ozone budget suggest that the transport of ozone from  
25 the stratosphere (approximately 500 Tg yr<sup>-1</sup>) is smaller than estimated in the TAR (770 Tg yr<sup>-1</sup>), while  
26 the photochemical production and destruction rates (5000 and 4500 Tg yr<sup>-1</sup>, respectively) are higher than  
27 estimated by TAR (approx. 3000 and 3500 Tg yr<sup>-1</sup>).
- 28 • Climate change affects air quality by modifying the dispersion rate of pollutants, the chemical  
29 environment for ozone and aerosol generation, and the strength of emissions from the biosphere, fires,  
30 and dust. Although the sign and magnitude of these effects are highly uncertain, and may vary greatly  
31 depending on region, there is the potential for significant air quality degradation.
- 32 • Long-term trends in the tropospheric concentration of OH, and hence in the oxidizing capacity of the  
33 atmosphere are determined by changes in the concentrations of methane, carbon monoxide and nitrogen  
34 oxides, and potentially by climate change. There are still large uncertainties in the model estimates and  
35 in the observation-derived values of the trends and inter-annual variability of the mean OH  
36 concentration. These changes have an important impact on the variability of the methane growth rate.
- 37 • Model calculations suggest that the effect on the OH burden of increasing concentrations of methane and  
38 carbon monoxide together with climate warming (which produces an increase in the methane oxidation  
39 rate) could be partly offset by increasing concentrations of NO<sub>x</sub> and water, so that future methane  
40 lifetime should remain relatively unchanged during the next decades.
- 41 • Model calculations suggest that past ozone depletion has induced surface cooling, which approximately  
42 balanced the greenhouse warming produced by increasing atmospheric abundances of ozone depleting  
43 substances and their substitutes.
- 44 • Since the pre-industrial era (circa 1750), the positive radiative forcing estimated from changes in  
45 tropospheric ozone has been larger than the negative forcing resulting from stratospheric ozone  
46 depletion. However, over the 1970–2000 period, these two forcings were of approximately equal  
47 importance, but opposite in sign.
- 48 • The changes in the stratospheric climate associated with the projected increase in the concentration of  
49 greenhouse gases could affect the recovery of polar ozone resulting from the phase-out of  
50 chlorofluorocarbons. This climate feedback should be more prominent in the Arctic than in the  
51 Antarctic.

### 52 *Aerosol Particles and Climate*

- 1 • An increasing aerosol load due to human activities decreases regional air quality and the amount of solar  
2 radiation reaching the earth's surface ("solar dimming"). In some areas such as eastern Europe, the  
3 "solar dimming" trend has been reversed since the collapse of the East Bloc.
- 4 • Increased light absorbing aerosol particles aloft, e.g., black carbon, lead to increasing atmospheric  
5 stability and a reduction in cloud amount, especially in polluted regions of the Northern Hemisphere.
- 6 • Increases in aerosol particles/clouds from human activities are causing a reduced daily range in surface  
7 temperature, with some evidence of a "weekend" effect due to differences in human activities on  
8 weekends and during the work week.
- 9 • Models suggest that global mean evaporation and precipitation have decreased in response to the  
10 increased aerosol load between pre-industrial times and the present, but projections indicate that both  
11 may increase with climate change over the next century.
- 12 • The magnitude of the overall indirect aerosol effect on clouds remains uncertain, because it comprises  
13 interactions with the climate system and feeds back on cloud and large-scale dynamics and the  
14 hydrological cycle.  
15

## 7.1 Introduction

The Earth system is complex. Nonlinear interactions involving physical, chemical and biological processes (Figure 7.1.1) occur either within specific compartments of the Earth system (atmosphere, ocean, land) or between them. Feedbacks can either be negative or positive. Negative feedbacks damp forcing applied to the system; in this case, they provide a stabilizing mechanism and tend to keep the Earth system close to its present equilibrium state. Positive feedbacks amplify applied perturbations, and potentially produce transitions towards a new equilibrium state. Thus, in the presence of positive feedbacks, minute actions could trigger abrupt, sometimes large and perhaps unmanageable changes in the Earth system.

[INSERT FIGURE 7.1.1 HERE]

An important aspect of climate research is to identify potential feedbacks, and determine if these tend to stabilize or destabilize the current climate system. In particular, it is important to assess if such feedbacks could produce large and undesired responses to perturbations applied as a result of human activities. Studies of past climate evolution on different timescales (including paleo-periods) can shed some light on the mechanisms that could trigger nonlinear responses to external forcing. The purpose of this chapter is to investigate some of the feedbacks of potential importance for the Earth system. Specifically, it will examine how terrestrial ecosystems, the carbon cycle, reactive gases and aerosol particles could affect the climate system. Large uncertainties remain in many issues discussed in this chapter, so that quantitative estimates of the importance of the coupling mechanisms discussed in the following sections are not always available. In addition, the regional disparity in the role of some cycles and the complex interactions between them limits our ability at present to provide a simple quantitative description of the interactions between biogeochemical processes and climate change.

### 7.1.1 Terrestrial Ecosystems and the Climate System

Far from being a passive recipient of changes in a coupled ocean-atmosphere system, the terrestrial biosphere interacts strongly with the climate, providing both positive and negative feedbacks due to biogeophysical and biogeochemical processes. Some of these feedbacks, at least on a regional basis, can be of the same order of magnitude as greenhouse gas forcing.

#### 7.1.4 Biogeophysical feedbacks

Changes in the structure or functioning of terrestrial ecosystems can feed back to climate via changes in evapotranspiration, albedo or roughness (which affects momentum transfer between the surface and the atmosphere). The evapotranspiration feedback is well-known. For example, half or more of the precipitation that falls on the Amazon Basin is recycled back to the atmosphere via evapotranspiration, affecting the climate both through cloud formation and through the partitioning of sensible and latent heat at the land surface; and the growing season is characterized by a small diurnal temperature range (Schwartz and Karl, 1990; Collatz et al., 2000; Durre and Wallace, 2001; Bonfils et al., 2004, 2005). Thus, changing the nature of terrestrial ecosystems through land-use change could feed back strongly on climate. Betts (2001) has estimated that past deforestation has led to decreases in near surface air temperature of up to 1°C over large areas of central North America and central Eurasia. This is about the same magnitude as the observed increase in surface temperature over the last century.

Terrestrial ecosystems are also coupled to climate via albedo feedbacks. The classic example is the so-called taiga (boreal forest) - tundra feedback system. The albedo of snow-covered vegetation is much lower for boreal forest than for tundra, as the snow lies underneath the dark canopy of the forest but over the top of the much lower tundra vegetation. The measured winter albedo differs sharply for the two vegetation types - about 0.75 for snow-covered tundra and 0.2 to 0.4 for boreal forest. The darker forest therefore absorbs more radiation than the bright, snow-covered tundra, warming the soil under the forest, which in turns favours the growth and expansion of the forest. This gives rise to the positive feedback effect. The taiga-tundra feedback effect has been shown to be important in the functioning of the Earth System in the past. The early to mid-Holocene climate in the northern high latitudes cannot be simulated correctly by considering orbital forcing and ocean-atmosphere dynamics only; the taiga-tundra feedback must be included (Berger, 2001).

#### 7.1.4.2 *Biogeochemical feedbacks*

These operate primarily through the carbon cycle, and there is a range of processes in terrestrial ecosystems that influence the flux of carbon between land and the atmosphere. Although there are many carbon compounds (e.g., CH<sub>4</sub>, VOCs) that terrestrial ecosystems exchange with the atmosphere, this brief summary will focus on processes that control the exchange of CO<sub>2</sub>:

The CO<sub>2</sub> fertilization effect. As atmospheric CO<sub>2</sub> concentration increases, photosynthetic rates of plants also increase. In general, this leads to higher carbon uptake by terrestrial vegetation but the relationship is nonlinear and is modified by several important interactions - with the hydrological and nutrient cycles, for example. The interreaction with the hydrological cycle is especially important through increases in water use efficiency due to increasing atmospheric CO<sub>2</sub> concentration, which further stimulates growth and uptake of carbon from the atmosphere. Also, different types of plants (e.g., those with C<sub>3</sub> and C<sub>4</sub> photosynthetic pathways) have intrinsically different responses to changing atmospheric CO<sub>2</sub> concentration. The CO<sub>2</sub> fertilisation effect is a negative feedback on increasing atmospheric CO<sub>2</sub> concentration and thus on a warming climate.

Nutrient mineralisation. Increasing temperature and changing moisture regimes have impacts on the mineralisation of nutrients in the soil, most notably on nitrogen compounds. This has the effect of releasing plant-available nitrogen compounds which act as a stimulant to growth, thus increasing the uptake by vegetation of carbon from the atmosphere. This is also a negative feedback on climate.

Heterotrophic respiration. Organic carbon compounds in the soil, originally derived from plant material, are slowly oxidised by bacteria and returned to the atmosphere as CO<sub>2</sub>. A variety of organic compounds are respired at different rates depending on the nature of the compound and on the communities of mycorrhizal fungi (Treseder et al., 2003). Both temperature and soil moisture can significantly affect rates of heterotrophic respiration, which generally increases with increasing soil temperature. Differences in the magnitude of this rate increase (or the lag between photosynthesis and respiration) were largely responsible for the spread in results of coupled carbon cycle-climate models, and the temperature-heterotrophic respiration relationship is an active area of research in terrestrial ecology. Changes in heterotrophic respiration with climate constitute a positive feedback.

Biome shifts. A shift of ecosystem structure can affect climate by changing the partitioning of carbon between the atmosphere and the land surface. These feedbacks can be either positive or negative, depending on the nature of the shift. For example, migration of boreal forest northward into tundra would initially lead to an increase in carbon storage in the ecosystem due to the larger biomass of trees than of herbs and shrubs. This would be a negative feedback. In the longer time frame (e.g., centuries), the changes in soil carbon would need to be considered to determine the overall effect. A shift in tropical rainforest to savanna, on the other hand (as simulated in the Cox et al., 2000 study), would result in a net flux of carbon from the land surface to the atmosphere and thus would be a positive feedback.

Disturbances. Fire is probably the most important disturbance in terms of the carbon cycle. Fire is a natural component of the dynamics of several biomes - for example, boreal forests and savannas. If averaged over a long period of time with no underlying change in frequency or extent, fires are neutral in terms of the carbon cycle; carbon lost to the atmosphere rapidly during a fire is regained slowly during the subsequent regrowth. However, if fires become more frequent or cover larger areas of land, such as appears to be happening in the Canadian boreal forests over the last several decades, such fire-affected biomes could become net sources of carbon to the atmosphere during the fire regime shift. This would be a positive feedback to climate, given that a warming climate increases the probability of fires.

Details on feedbacks between climate and the land surface are provided in Section 7.2.

#### 7.1.2. *The Carbon Cycle and the Climate System*

During the last decade, the scientific community has realized that interactions between biogeochemical processes and the physical climate system could generate additional feedback processes, and hence affect the future evolution of the Earth's climate. An important example is provided by the climate carbon interactions. In the past, models treated the changes in the atmospheric CO<sub>2</sub> concentration as an external forcing factor,

1 and derived the corresponding response of temperature. Recently, however, it was recognized that the level  
2 of atmospheric CO<sub>2</sub>, which increases with the intensity of fossil fuel emissions, also depends on the rate of  
3 carbon uptake by the ocean and by the land, and that these uptake fluxes are themselves dependent on the  
4 atmospheric CO<sub>2</sub> level and on climate.  
5

6 The dynamics of carbon cycle-climate interactions through this century have been simulated using coupled  
7 global carbon-climate models (Friedlingstein et al., submitted). These models suggest that the overall effect  
8 of carbon cycle-climate interaction is a positive feedback; by 2100 the atmospheric CO<sub>2</sub> concentration is  
9 higher and the climate is consequently warmer in all coupled model runs than for the uncoupled runs.  
10 Differences in the magnitudes of the feedbacks among the models, which remain substantial, are due to  
11 different underlying sensitivity associated with the representation of both climate and carbon cycle processes  
12 in the models. More details on this issue are provided in Section 7.3.  
13

### 14 *7.1.3. Reactive Gases and the Climate System*

15

16 Interactions between climate and atmospheric oxidants, including ozone, provide other important coupling  
17 mechanisms in the Earth system. The concentration of ozone has increased substantially in the troposphere  
18 since the pre-industrial era, and has contributed to radiative forcing, especially in polluted areas of the world.  
19 Below the tropopause, ozone is produced by photo-oxidation of carbon monoxide (CO), methane (CH<sub>4</sub>) and  
20 nonmethane hydrocarbons in the presence of sufficiently high levels of nitrogen oxides (NO<sub>x</sub>). The  
21 emissions of ozone precursors have increased as a result of larger use of fossil fuel, more frequent biomass  
22 burning and more intense agricultural practices. The level of pre-industrial ozone is not accurately known, so  
23 that the resulting radiative forcing cannot be accurately determined, and must rely on model estimates.  
24

25 What are even less well quantified are the changes in the atmospheric composition that could result from  
26 climate changes, and could involve different processes. The photochemical production of the hydroxyl (OH)  
27 radical, which occurs in the presence of ozone and water vapour, should be enhanced in a wetter atmosphere.  
28 Since OH destroys ozone, this climate chemistry feedback should be negative, and should limit the future  
29 increase in tropospheric ozone that will result from enhanced anthropogenic emissions of the precursors. At  
30 the same time, the atmospheric concentration of methane, a greenhouse gas whose main loss is due to the  
31 reaction with OH, should also be reduced, so that here again, the feedback is negative.  
32

33 Other chemistry-related processes could provide an amplification of the initial effect. For example, if the  
34 frequency of lightning flashes increases in response of climate warming, the nitrogen oxides released by  
35 thunderstorms would lead to additional ozone production, and hence to enhanced greenhouse warming. The  
36 effect on ozone of a simultaneous increase in NO<sub>x</sub> and water vapor is complex because hydroxyl radicals  
37 produced by water modify the partitioning between nitrogen compounds. The scavenging mechanisms  
38 affecting several chemical species will also be modified in response to climate-driven perturbations in the  
39 hydrological cycle. Changes in the natural emissions of chemical compounds (e.g., biogenic hydrocarbons  
40 by the vegetation, nitrous and nitric oxide by soils, ocean exchanges of chemical compounds) in response to  
41 climate change provide the potential for additional feedback mechanisms. The importance of these effects is  
42 not yet well quantified. Several of them depend on the expected evolution in the land vegetation as climate  
43 evolves. These issues are further discussed in Section 7.4.  
44

45 Finally, the recovery of stratospheric ozone in the future decades could also be affected by future climate  
46 changes. Significant amounts of ozone are destroyed by chlorine compounds, which are released in the  
47 stratosphere by industrially manufactured chlorofluorocarbons. In particular, the chemical destruction of  
48 ozone at high latitudes requires the presence of polar stratospheric cloud (PSC) particles, on which chlorine  
49 is activated by heterogeneous chemical processes. The formation of these clouds is a strong function of  
50 temperature and of the water vapour concentration. A cooling of the lower polar stratosphere, caused by  
51 enhanced radiative cooling by CO<sub>2</sub>, could facilitate the formation of PSCs, produce more ozone destruction,  
52 and delay the ozone recovery. If this process is accompanied by an intensification of the Brewer-Dobson  
53 meridional circulation, a warming of the polar region could occur, with a reduction in the chemical ozone  
54 destruction, and probably an acceleration of the ozone recovery. This question is also discussed in Section  
55 7.4.  
56

#### 7.1.4 *Aerosol Particles and the Climate System*

The presence of tiny sulphate aerosol (SO<sub>4</sub>) particles in the atmosphere tends to cool the Earth's surface by scattering some of the incoming solar radiation back to space. In addition, by acting as condensation nuclei (CCN), these particles tend to modify the radiative properties of the clouds (Twomey effect) or their lifetime (Albrecht effect), which contribute to additional surface cooling. As the major natural source of sulphate is provided by dimethyl sulphate (DMS), an organic compound whose production in the ocean and release to the atmosphere depends on climatic factors, and specifically on the temperature of the upper ocean, Charlson et al. (1987) suggested that the sulphur cycle could generate a feedback loop whose sign and amplitude remain uncertain. In many areas of the Earth, however, the importance of this potential feedback loop has been considerably reduced by the presence of large amounts of SO<sub>4</sub> particles produced as a result of human activities (e.g., coal burning). With an elevated atmospheric aerosol load, principally in the Northern hemisphere, it is likely that the temperature increase during the last decade has been smaller than the increase that would have resulted from the climate forcing by greenhouse gases alone. Other indirect effects of aerosols on climate have been suggested, including the evaporation of cloud particles through absorption of solar radiation by soot, which in this case, provides a positive warming effect. These questions are discussed in Section 7.5.

Aerosols deriving from human activity are also delivering more nitrogen and iron to the surface ocean (e.g., Tegen et al., 2004). These nutrients are 'new' to the ocean (Meshkhidze et al., 2003), therefore having the potential to increase the export of biogenic carbon to the ocean interior, and they also have the potential to act as ballast, also increasing the sinking of biogenic carbon to the ocean interior (e.g., Klaas and Archer, 2002). Both effects represent a negative feedback to atmospheric CO<sub>2</sub> concentrations.

### 7.2 **The Changing Land Surface and Climate**

#### 7.2.1 *Introduction to Land Climate Response from Human Activities*

Land surface as a dynamical component of the climate system (illustrated in Figure 7.2.1) responds to inputs from the atmosphere, including radiation, precipitation, and other meteorological variables of the overlying boundary layer. How it responds to these inputs is controlled by its structure, i.e., all the details of its governing processes. Its structure may involve how it exchanges moisture and heat with the atmosphere or what is its vegetation state and how that is determined by slow biological processes. There is a large variety in the processes included in land models and in the concepts that are used to express them mathematically. Furthermore, this structure is modified by both climate change and land use change. Thus, the outputs from a land model depend on a wide variety of factors and so are not easily evaluated in terms of their reality.

[INSERT FIGURE 7.2.1 HERE]

Humans influence the physical land climate through modifications of atmospheric greenhouse gas and aerosol concentrations and by land use change. The consequent changes in land climate over long time scales can modify the land cover state with similar consequences as anthropogenic land use change. The natural and anthropogenic land cover changes also affect the exchange of carbon between the land and the atmosphere. Changes in the physical land climate can also modify land-atmosphere carbon exchanges. What is most important depends on the question being addressed: e.g., what is the a) change of surface land climate in response to changing global radiative balances from greenhouse gases; b) feedbacks between land and atmospheric variability; c) bias in simulation of current climate; d) responses of surface land climate to changing land use? Although a) may be the primary focus of AR4, all these questions have been addressed in recent literature and collectively are important for determining the interactions between land climate changes and human activities.

#### 7.2.2 *How Does Land Surface Mediate Climate Change?*

##### 7.2.2.1 *Why does land have a larger effect on regional than global climate?*

On a global spatial scale and longer time scales, climate change is largely controlled by processes of global radiation balance. Land is only about 30% of the globe, and its ability alone to affect global radiation balance is largely through its ice and snow cover, and the shading of the latter by vegetation. Processes such as desertification that make the surface reflect more radiation (i.e., increase its albedo) generally occur over

1 relatively small areas and are judged to collectively have a relatively small effect. However, on a regional  
2 scale and at the surface, many additional more localized and shorter time scale processes can affect climate  
3 in other ways, and possibly be of comparable or greater importance. Variability in tropical ocean  
4 temperatures, e.g., connected to ENSO, determines major changes in the patterns of precipitation and  
5 drought over continents

6  
7 Shifts in cloudiness can occur and atmospheric circulation systems can redistribute energy to maintain  
8 overall balances. Because low clouds are normally closely coupled to the surface, those over land can be  
9 significantly changed by changes of land properties. Land has a strong control on the vertical distribution of  
10 atmospheric heating, through its Bowen ratio and radiative exchanges (e.g., Betts, 2004). The Bowen ratio  
11 determines how much of the radiation delivered to land goes into the near surface boundary layer versus how  
12 much is delivered as latent heat at higher levels. Land is more sensitive to changes in its energy balance  
13 under cold stable conditions than under warm unstable ones because of the dependence on stability of  
14 surface exchange coefficients. Consequently, a change in the diurnal amplitude of surface radiative heating  
15 can have a substantial impact on night-time and hence average temperatures. If applied locally,  
16 compensating effects may be expected elsewhere. Effects that modify local or regional temperatures may  
17 also influence distributions of precipitation, but this possibility is difficult to assess because of model  
18 differences and an inadequacy of quantitative studies.

19  
20 Changes over land that modify the Bowen ratio can cause large changes of surface temperature, both locally  
21 and regionally. These changes will produce adjustments elsewhere to maintain global energy balance.  
22 Attempts have been made to find such remote adjustments (Avisar and Werth, 2005). Such adjustments  
23 occur in multiple ways, and are part of the dynamics of climate models. For example, the locally warmer  
24 temperatures can lead to more rapid vertical decreases of atmospheric temperature so that at some level  
25 overlying temperature is lower and radiates less. The net effect of such compensations is that averages over  
26 larger areas will give much smaller estimates of change.

27  
28 Thus such regional changes are better described by local and regional metrics or on larger scales by  
29 measures of change in spatial and temporal variability rather than simply in terms of a mean global quantity.  
30 The large amount of such details involved is best addressed by assessments at a local and regional scale.  
31 However, the IPCC with its global perspective should provide overall guidance regarding these issues and  
32 how well they can currently be quantified and how they couple to global climate change.

#### 33 34 *7.2.2.2 What has been learned regarding forcing terms important for land climate?*

35 Local precipitation intensities are increasingly considered in some ways to be as important as precipitation  
36 amounts. However, it is difficult for climate models to quantify such intensities. A related dimension is the  
37 amplitude and phase of the diurnal cycle in precipitation, which is simulated by climate models but  
38 apparently not yet very well (e.g., Collier and Bowman, 2004). Betts (2004) reviews how the diurnal cycle of  
39 tropical continental precipitation is linked to land surface fluxes and argues that errors in a model can feed  
40 back on model dynamics with global impacts.

#### 41 42 *7.2.2.3 How is land structure characterized?*

43 Because of its complexity, there are no uniformly accepted approaches to including land structure in a  
44 climate model. It has been treated with a wide variety of approaches. Table 7.2.1 shows a possible  
45 breakdown into various structural components and an assessment of their relative importance and  
46 understanding from the viewpoint of inclusion in a climate model. This is derived from consideration of the  
47 literature since TAR (reviewed here) as well as earlier work.

48  
49 Simulation of annual runoff is reported by Milly and Shmakin (2002a) to be primarily sensitive to values  
50 assumed for aerodynamic roughness and the leaf stomatal resistance. They identified a major bias in regions  
51 of seasonal dryness that they attributed to their lack of accounting for two-way coupling between their root-  
52 zone and deeper aquifer water storage. They found a dry bias in modelled evapotranspiration, which they  
53 attributed to several factors but in particular, their neglect of leaf water evaporation. Milly and Shmakin  
54 (2002b) found, as might be expected, a substantial improvement in their modelled runoff from use of a  
55 vegetation type-based stomatal conductance varying geographically rather than a global value. Some features  
56 such as treatment of leaf water evaporation may have little effect on the determination of monthly  
57 evapotranspiration (e.g., as found in the analysis of Desborough, 1999) but still produce important changes

1 of temperature and precipitation. Much of the model structure is intended to address short time scale and  
 2 diurnal variability and its potential coupling to the atmosphere. For example, Wang and Eltahir (2000)  
 3 compare model results (their Figure 1) for realistic versus uniform precipitation intensities. Both cases have  
 4 about the same evapotranspiration, but evidently, for the latter case, the feedback on precipitation is so  
 5 strong that their runoff drops from 700 mm y<sup>-1</sup> to nearly zero. Other recent studies such as reviewed here  
 6 also suggest that identical scenarios for global warming or land-use change introduced into models with  
 7 different structures may give significantly different outputs of land climate. On the other hand, Pitman et al.  
 8 (2005) in a coupled study with land configurations of different complexity and forced by AMIP II SST's  
 9 were unable to find any impacts on atmospheric variability.

10  
 11 **Table 7.2.1.** Preliminary assessment of structural factors important for the land response to global climate  
 12 change and land use change. Current literature addressing these questions is fragmented and conclusions are  
 13 likely to be dependent on both land and atmospheric models used.  
 14

Structural Factor	How Important?	Certainty of its Modelling
Roughness	***	***
Bowen ratio (depends on collective effect of several other structural factors)	***	**
Soil moisture Profile	**	**
Root representation	*?	*
Complex micrometeorology	x??	x
albedo	**	**
Canopy/stomatal conductance	*?	*
Partitioning of radiation	*?	*

15 Notes:

16 \*\*\* = high

17 \*\* = moderate

18 \* = low

19 x = very low

20 ? = not enough evidence but could add another \*

21  
 22  
 23 Much of the current literature addressing the issue of the appropriate structure to use for land processes in  
 24 climate models has been asking what is the impact of additions to or observational improvements of  
 25 vegetation complexity. When this additional complexity incorporates general knowledge or new  
 26 observational constraints, it might be thought of as an improvement to a model. Whether it is useful enough  
 27 to be adopted by others then depends on the answer to the question “what difference does it make?” There  
 28 has been a considerable body of literature reporting on the consequences for various climate models of  
 29 improvements in their land models of their descriptions of leaf cover and descriptions of types of vegetation  
 30 cover. Henderson-Sellers (2003) in comparing the surface fluxes among 19 models submitted to the AMIP II  
 31 intercomparison, reports for controlled conditions, over an order of magnitude difference between sensible  
 32 fluxes of different models. However, more recently developed models cluster much more tightly.  
 33

34 Many modelling studies have been able to demonstrate that changing land cover can have local and regional  
 35 climate impacts that are comparable in magnitude to temperature and precipitation changes observed over  
 36 the last several decades as reported in Chapter 3. Current literature demonstrates large disparities in  
 37 conclusions. For example, Snyder et al., (2004) show that removal of temperate forests can give a summer  
 38 (JJA) warming of 1.3°C and a reduction in precipitation of -1.5 mm day<sup>-1</sup>. Oleson et al., (2004) on the other  
 39 hand, conclude that removal of temperate forests in the US would cool JJA by 0.4 to 1.5°C and probably  
 40 increase precipitation depending on details of the model and prescription of vegetation.  
 41

42 The discrepancy between these two studies published in the same edition of the same journal, may in this  
 43 case be largely an artifact of visibly different assumptions as to the endpoint model structure. The first study  
 44 assumes conversion of forest to desert and the second to crops. However, the large number of such studies  
 45 that demonstrate a potentially important impact of human activities on climate through land use modification  
 46 suggest that IPCC should incorporate in its assessments of regional climate change the possible contributions  
 47 of land use and land cover change.

### 7.2.3 What Has Been Learned About Land Model Structures Since the TAR?

#### 7.2.3.1 Properties affecting radiation

*Albedo and emissivity have long been recognized as a land contribution to radiative balance.*

Surfaces that have more or taller vegetation are commonly darker than those with short or sparse vegetation. With sparse vegetation, the net surface albedo also depends on albedo of the underlying surfaces, especially if a light soil or snow. For models that separately balance canopy and surface energy budgets, the partitioning of radiative fluxes between these components also becomes important.

New satellite data show the importance of radiation heterogeneities on the plot scale for the determination of albedo and PAR, and appropriate modelling concepts are being advanced (e.g., Yang and Friedl (2003) and Niu and Yang, 2004; Pinty et al., 2005). The inclusion of reasonably accurate values of emissivity in models has been addressed by Zhou et al. (2003a,b) and Ogawa and Schmugge (2004).

#### 7.2.3.2 Properties affecting Bowen ratio

Soil moisture control of the partitioning of that energy between sensible and latent flux has been demonstrated to be very important for local and regional temperatures, and their coupling to precipitation. Oglesby et al. (2002) carried out an initially dry soil anomaly study where the dryness of the soil over the US Great Plains for at least the first several summer months of their integration produces a warming of about 10–20°C. The simulated mass flux from moist convection has been demonstrated important for surface fluxes. In particular Williamson et al. (2005), using data from the ARM study site, have shown that flaws in model parameterizations of convection can cause perturbations in evapotranspiration that lower temperatures by more than 1°C from excessive evapotranspiration.

How can vegetation modify the Bowen ratio? The most important factors for Bowen ratio (e.g., Table 7.2.1) are surface roughness, leaf area, and availability of water from the soil for use of vegetation. The height of the vegetation is the most important determinant of surface roughness that determines the production of mechanical turbulence. Whether water has been intercepted on the surface of the leaves or its loss is only from the leaf interior as controlled by stomates makes a large difference. Vegetation that is shorter and with more leaves has the most latent flux and the least sensible flux. A replacement of forests with shorter vegetation together with the normally assumed higher albedo should then cool the surface. However, if the replacement vegetation has much less foliage or cannot access soil water as successfully, a warming may occur. Thus deforestation can modify surface temperatures by up to several degrees in either direction depending on the details of what type of vegetation replaces the forest and the climate regime assumed. Drier air can act to increase evapotranspiration but leaves may have negative feedbacks through their conductance to reduce this effect. In absence of leaves, forests in early spring appear as especially dry surfaces with consequent large sensible fluxes and high boundary layer (e.g., Betts et al., 2001)

Representation of leaf phenology in models has advanced from specified constant leaf cover or simple phenology rules to prescribed leaf cover (e.g., van den Hurk et al., 2003; Buermann et al., 2001) that is constrained by observational estimates (e.g., Tian et al., 2004a-c) and to prognostic approaches that estimate leaf cover on the basis of physiological processes (e.g., Arora and Boer, 2005a). Arora (2005) gives a brief overview of the issues in modelling stomatal conductance for climate simulations. Levis and Bonan (2004) discuss how in middle latitude forests the springtime leaf emergence when it is interactively modelled provides a negative feedback on the rapid increases of temperature by their increased transpiration.

Addressing the dynamics of leaf carbon uptake as coupled to evapotranspiration may require consideration of the controls by nitrogen and the nutrient cycling (e.g., Dickinson et al., 2002). The difference in temperature of leaves in sun or shade has a significant effect on canopy transpiration (Dai et al., 2004). Dynamic vegetation models have advanced and explicitly simulate competition between plant functional types (PFTs) (Sitch et al., 2003; Bonan et al., 2002a, b, 2003), e.g., Arora and Boer (2005b) suggest a generalized form of competition-colonization equations that permit equilibrium coexistence of different PFTs.

Surface fluxes interact with the boundary layer and lead to various additional model feedbacks (depending on details of other modelling components) involving changes of humidity, boundary-layer heights, cloud-

1 fraction and precipitation, e.g., Ek and Holtslag (2004) discuss conditions under which reduced  
2 evapotranspiration can increase boundary layer cloudiness.

### 3 4 *7.2.3.3 Detailed vertical layering*

5 The importance of simulating resolved vertical profiles of soil moisture and temperature (Rosnay, 2003; Dai,  
6 2003) and hydrological effects of horizontal heterogeneity of topography (Koster et al., 2000) has been  
7 emphasized in recent work. The parameterization of water uptake by roots contributes to the computed soil  
8 water profile (Feddes et al., 2001; Barlage and Zeng, 2004), and efforts are being made to make the roots  
9 interactive, e.g. Arora and Boer (2003). Wu and Dickinson (2004), and Wu et al. (2002a, b) characterize the  
10 coupling of layered soil water to ET and climate variability. Models have also moved from single slab to  
11 multilayer snow models (Stieglitz et al., 2001; Dai et al., 2003).

### 12 13 *7.2.3.4 Recognizing spatial complexities*

14 Various heterogeneities of snow distributions have been shown to be important for determining the melting  
15 of snow and other aspects of surface energy balances, but these are not yet considered in global climate  
16 models. Approaches to use of a subgrid distribution of snow properties have been addressed by Roesch et al.  
17 (2001) and Liston (2004); and snow depth modelling by Strack et al. (2004). Sturm et al. (2001) describe  
18 how Arctic shrubs promote snow depth and how warming would increase snow depths.

19  
20 Energy exchanges between surface and canopy are important for overall surface temperature determination  
21 (e.g., Dickinson et al., 2005; Zeng et al., 2005) and its remote sensing (Friedl, 2001). Modelling of  
22 turbulence of complex canopies has been advancing in directions that can provide improved formulations  
23 (e.g., Katul et al., 2004; Poggi et al., 2004).

### 24 25 *7.2.3.5 Can increased complexity make any difference?*

26 Many details such as described above may be nearly invisible in comparisons with the more standard  
27 meteorological measurements. Thus they may be of little interest for models whose only metric is a matching  
28 to such measurements. In some cases, they may be simply a response to speculation that such further details  
29 will turn out in some way to be beneficial to a model. However, they are mostly driven by important  
30 modelling questions that cannot be answered without inclusion of such complexity or by the data and  
31 important questions of other communities at smaller spatial scales. For example, there is considerable  
32 literature advancing the hypothesis that the presence of vegetation will provide a positive feedback on the  
33 melting of snow through its lower albedo. This hypothesis has been supported by models that assumed that  
34 energy absorbed by the vegetation would be delivered to the snow. This is not obvious and its demonstration  
35 by a climate model requires the inclusion of more complex micrometeorology and radiative transfer than  
36 implemented in current models.

37  
38 Greater complexity in hydrological structures of climate models is commonly suggested by advances in  
39 understanding of surface hydrology and of plot level soil and vegetation descriptions by ecological  
40 measurements. For example, the need for detailed layering in snow was discovered by soil temperature and  
41 respiration measurements made in the BOREAS field program. The previous single slab snow models  
42 generated soil temperatures tens of degrees too cold, although this error had little direct effect on surface air  
43 temperatures. Climate models are expected by various communities to provide relevant information for other  
44 parts of the system than simply the atmosphere, which can only be achieved if the models include adequate  
45 complexity to describe the processes of interest.

46  
47 It is always possible that improvements expected from adding structure to land models may be masked by  
48 remaining errors common to several or all models in their atmospheric simulations. Gutowski et al. (2003)  
49 analyze such a dry bias common to several models in looking at precipitation in the South Central US.

## 50 51 **7.2.4 How Can We Quantitatively Relate Land Climate Changes to Atmospheric Inputs and Structural** 52 **Change?**

### 53 54 *7.2.4.1 Assessing quantitative sensitivity is complicated*

55 Temperature change on a global scale is largely related to changes of global radiative forcing at the top of  
56 the atmosphere. However, changes of near surface temperatures on regional or smaller scales may be locally  
57 of equal significance and occur for reasons other than global radiative forcing. Structural changes from land

1 use change or changes in precipitation or in radiative exchanges between surface and atmosphere may drive  
2 important land climate change without any connection to global radiative forcing.

3  
4 Such changes, however, are more poorly understood than those forced by global radiation. Sensitivity should  
5 be considered as a multivariate issue. It includes the question as to how near-surface temperature (which  
6 more directly affects human activities) can change independently of the “global temperature” and how  
7 “wetness” can change. A difference between atmosphere and surface temperature can result from a thermal-  
8 radiative decoupling (e.g., Betts et al., 2004).

9  
10 “Wetness” describes the strong Bowen ratio controls on surface fluxes. The presence of vegetation  
11 complicates the concept of surface wetness in ways not yet accounted for in all global models. Wetness in  
12 the presence of vegetation involves water stores on leaves, in shallow soil and in deep soil. The latter has  
13 received most attention in discussion of evapotranspiration. It represents the bulk of the water stored and  
14 imposes the longest time scale. However, it is the rapidity of evaporation of the near surface stores that  
15 allows them to be of comparable importance for surface water and energy balances. (Dickinson et al, 2003  
16 discuss the significance of these disparate timescales.) On longer time scales, evaporation from the fast  
17 reservoirs acts primarily as a surface energy removal mechanism, and from the water viewpoint, largely to  
18 cancel some of the incident precipitation. Another complication, currently not included very well in climate  
19 models, is that over some fraction of moist soils, water tables can be high enough to be hydrologically  
20 connected to the rooting zone, or even to the near surface store as in wetlands (e.g., Koster et al. (2000),  
21 Marani et al., 2001; Milly and Shmakin, 2002a; Liang et al., 2003; Gedney and Cox, 2003).

22  
23 The study by Scanlon et al. (2005) provides a nice example of how the dynamics of wetness, expressed as  
24 soil moisture, can depend on vegetation. She monitored soil moisture in the Nevada desert with lysimeters  
25 either including or excluding vegetation and for a multiyear period that included times of anomalously strong  
26 precipitation. Without vegetation, much of the moisture penetrated deeply, had a long lifetime and became  
27 available for recharge of deep groundwater, whereas for the vegetated plot, the soil moisture was all  
28 extracted by the plants for transpiration.

29  
30 Climate model simulations of the climate change deriving from greenhouse gases include significant  
31 feedbacks between the land surface and atmospheric precipitation. Some observational evidence for such  
32 feedback has been noted by D’Odorico and Porporato (2004) in support of a low-dimensional model of such  
33 feedbacks. Analysis of such feedbacks in full climate models is difficult: only recently have efforts been  
34 initiated to examine this issue (Koster et al., 2004). Unfortunately, the initial conclusion is that from a  
35 statistical viewpoint, this feedback is poorly constrained, with some models having strong coupling from soil  
36 moisture back to precipitation, and some almost none. Such a coupling may be an important component of  
37 the description of regional climate change resulting from global warming or land use change, and it  
38 determines the potential predictability of precipitation from modelling of soil moisture.

#### 39 7.2.4.2 *Mechanisms for forcing the diurnal temperature cycle*

40 The diurnal cycle of temperature over land is maintained by daytime solar heating and nighttime radiative  
41 cooling. Daytime warm season heating produces a thick convective boundary layer with substantial heat  
42 capacity which is consequently insensitive to perturbations in the amplitude of the diurnal radiative forcing.  
43 Thus, daytime warm season temperatures are most readily changed by changing the Bowen ratio. Such  
44 changes may occur from changes of land structure such as vegetation cover, e.g., Bonan (2001), or by  
45 changes in precipitation. Nighttime and high latitude winter surface temperatures, on the other hand, are  
46 most readily altered by changes in atmospheric downward thermal radiation.

47  
48  
49 Qian and Giorgi (2000) discussed how the rapid growth of SO<sub>2</sub> emission in China since 1979 should be  
50 producing regional aerosol effects, and noted a reduction of the day-night temperature range of –  
51 0.26°C/decade over Sichuan. For reasons mentioned above this is difficult to attribute directly to reduction  
52 of solar heating. Bonan (2001) and Oleson et al. (2004) indicate that conversion of forests to agriculture  
53 could give such a daytime cooling. However, changes of land use of the magnitude required to give such a  
54 result seem unlikely. Huang et al. (2005) have modelled the growth of sulphate aerosols and their  
55 interactions with clouds in the context of a regional climate model, and find over Southern China a decrease  
56 in the day-night temperature range that is comparable with that observed by Zhou et al.(2004) and Qian and

1 Giorgi. The analysis of Huang et al. shows the nighttime temperature to be a result of increased nighttime  
2 cloudiness connected to the increase of aerosols.

#### 3 4 *7.2.4.3 Mechanisms for forcing climate change from spatial heterogeneity*

5 Clark et al. (2004) show an example of a squall-line simulation where variation of soil-moisture on the scale  
6 of the convection substantially modifies the rainfall pattern. Pielke (2001), Weaver et al. (2002), and Roy et  
7 al. (2003) have also addressed various aspects of convective scale precipitation coupling to land surface  
8 heterogeneity. If deforestation occurs in patches rather than uniformly, the consequences for precipitation  
9 could be quite different. Avissar et al. (2002) suggest that there may be an increase in precipitation resulting  
10 from partial deforestation as a consequence of the mesoscale circulations triggered by the deforestation, as in  
11 Silva Dias et al. (2002b). Such studies have indicated mechanisms by which small scale inhomogeneities  
12 may modify precipitation. However, characterizations of how land has changed globally to force such  
13 modifications are not yet available.

#### 14 15 *7.2.4.4 Sensitivity to vegetation change*

16 Maynard and Royer (2004a) address the sensitivity to different parameter changes in African deforestation  
17 experiments and find that changes of roughness, soil depth, vegetation cover, stomatal resistance, albedo,  
18 and leaf area index all could make significant contributions. Voltaire and Royer (2004) find that such  
19 changes may impact temperature and precipitation extremes more than means, in particular the daytime  
20 maximum temperature and the drying and temperature responses associated with El Niño events. Snyder et  
21 al. (2004a,b) also address the response to vegetation removal and African deforestation.

22  
23 Several studies have linked changes of land use to the climate change expected from increasing greenhouse  
24 gases. Maynard and Royer (2004b) find that anticipated changes in land cover modify the response of  
25 African climate to that of the greenhouse warming, in particular by further increasing the temperatures.

26  
27 Guillevic et al. (2004) address the issue of the importance of interannual variability of leaf area as inferred  
28 from AVHRR satellite data, and conclude that substantial sensitivity exists. In contrast, Lawrence and Slingo  
29 (2004) find very little difference in climate simulations between use of annual mean value of vegetation  
30 characteristics versus a prescribed seasonal cycle. They indicate some scepticism as to the realism of their  
31 result and suggest some model modifications that would give a much larger sensitivity.

32  
33 Osborne et al. (2004) examine effects of changing tropical soils and vegetation: variations in vegetation  
34 produce variability in surface fluxes and their coupling to precipitation. Thus, interactive vegetation can  
35 promote additional surface and atmospheric variability as analysed by Crucifix et al. (2005).

#### 36 37 *7.2.4.5 How can regional scale changes of vegetation modify precipitation regimes?*

38 Marengo and Nobre (2001) found that removal of vegetation led to a decrease in precipitation and  
39 evapotranspiration and a decrease in moisture convergence in central and northern Amazonia. Oyama and  
40 Nobre (2004) show how removal of vegetation in northeastern Brazil would substantially decrease  
41 precipitation.

42  
43 New developments in dynamic vegetation schemes and coupled climate-carbon models (Cox et al., 2000;  
44 Betts et al., 2003; Huntingford et al., 2004) have demonstrated the possibility of large feedbacks between  
45 further climate change and vegetation change. In particular, they found a large die-back in the Amazon  
46 vegetation and large reductions in Amazon precipitation. They also showed that the physiological forcing of  
47 stomatal closure from the rising CO<sub>2</sub> levels could contribute 20% to that rainfall reduction. Their simulated  
48 forest die-back also exerts two positive feedbacks on the precipitation reduction: (1) a biogeophysical  
49 feedback through reduced forest cover suppression of local evaporative water recycling, and (2) a  
50 biogeochemical feedback through the release of CO<sub>2</sub> contributing to an accelerated global warming (Betts et  
51 al., 2003). Levis et al., (2004) demonstrate a dynamic coupling between changes of African rainfall and  
52 vegetation.

53  
54 How land cover and its change help structure rainfall transitions has been considered by Fu and Li (2004)  
55 and DeFries et al. (2002b). Grimm (2003) has examined a possible feedback between soil moisture and the  
56 monsoon activity over tropical South America

1 7.2.4.6 *How can changes of land properties change boundary layer properties to modify occurrence of*  
2 *clouds and their radiative effects?*

3 Ek and Holtzlag (2004) discuss soil moisture cloud feedbacks as already mentioned. Chagnon et al. (2004)  
4 present strong observational evidence of a large increase in boundary layer clouds in the Amazon in areas of  
5 partial deforestation.

6  
7 The triggering of the annual South American Monsoon System may be connected to (1) a loading of aerosols  
8 due to biomass burning and (2) consequent modification of solar radiation (Artaxo et al., 1990) and  
9 atmospheric thermodynamic stability. Recent model and observational results have indicated that the aerosol  
10 plume produced by biomass burning at the end of the dry season is transported to the south and may interact  
11 with frontal systems, thus indicating a possible feedback to the precipitation regime (Freitas et al., 2005)  
12 through the radiative of cloud microphysical processes (Silva Dias et al., 2002).

13  
14 7.2.4.7 *How can the presence of aerosols and clouds modify land properties such as their carbon storage?*

15 Aerosols and clouds reduce the availability of visible light needed by plants for photosynthesis. However,  
16 the leaves in full sun may be light-saturated, i.e. they do not develop sufficient enzymes to utilize that level  
17 of light. Leaves that are shaded, however, are generally light limited. They are only illuminated by diffuse  
18 light scattered by overlying leaves, or by atmospheric constituents. Thus, an increase of diffuse light at the  
19 expense of direct light will promote leaf carbon assimilation and transpiration. Because of the observational  
20 uncertainties as to whether or not the additional diffuse light can overcompensate for the greater loss of  
21 direct light, this topic is controversial. It has been addressed by Gu (2002, 2003); Roderick et al., 2001;  
22 Cohan et al., (2002). That the aerosol-induced increase in diffuse radiation by the Mt. Pinatubo eruption  
23 could provide an enhanced terrestrial carbon sink has been suggested as an explanation of the temporary  
24 decline in the growth rate of atmospheric CO<sub>2</sub> that followed the eruption (Roderick et al., 2001; Gu et al.,  
25 2003).

26  
27 7.2.4.8 *How can vegetation structure be changed by changing climate?*

28 Analyses of satellite sensed vegetation greenness and meteorological station data suggest an enhanced plant  
29 growth and lengthened growing season duration in northern high latitudes since the 1980s (Zhou et al., 2001,  
30 2003c). This effect is further supported by modelling linked to observed climate data (Lucht et al., 2002).  
31 Nemani et al. (2002, 2003) suggest that increased rainfall and humidity spurred plant growth in the United  
32 States and changes in climate have eased several critical climatic constraints to plant growth and thus  
33 increased global terrestrial net primary production.

34  
35 7.2.5 ***Functioning of System Models - Improving Their Basis for Quantitative Evaluation of Impacts of***  
36 ***Land Changes***

37  
38 7.2.5.1 *Lower complexity model characterizations*

39 Because full realistic coupled climate models are so complex, analyses of the various potential feedbacks  
40 have been rather limited. Thus, potentially important mechanisms are better initially described in low or  
41 intermediate complexity models. Lower complexity models have been developed of interactive vegetation  
42 with multiple equilibria (Zeng et al., 2002; Foley et al., 2003; Wang et al., 2004; Zeng et al., 2004). Such  
43 models demonstrate how assumed feedbacks between precipitation and surface fluxes generated by dynamic  
44 vegetation may lead to the possibility of flip-flops between two soil moisture and precipitation regimes. The  
45 Sahel margin appears to most readily generate alternating precipitation regimes.

46  
47 7.2.5.2 *Model feedback intercomparisons*

48 Because of the complicated nature of couplings between land and the atmospheric hydrological cycle, their  
49 presence in a model gives no assurance of their correctness. Observational validation however, can be  
50 difficult to achieve. An alternate likely more limited, but potentially more quantitative evaluation approach,  
51 is the intercomparison of models from multiple groups. Land fluxes alone have been so intercompared for a  
52 number of years under the auspices of Project for Intercomparison of Land Surface Schemes (PILPS). The  
53 important question of how models differ in their coupling to the atmosphere has been less thoroughly  
54 addressed. Irannejad et al. (2003) have developed a statistical methodology to fit monthly fluxes from AMIP  
55 II climate models to a simple linear statistical model, depending on such factors as monthly net radiation,  
56 and surface relative humidity. With this fitting, they are able to run any such model with the forcing from  
57 any other model and in this way are able to characterize contributions to flux differences from the

1 atmospheric forcing versus the surface model. Apparently both are major source of variability; they find that  
2 the coupled models are more in agreement because of offsetting differences in the atmospheric and land  
3 models.

4  
5 It has long been reported in individual modelling studies that soil moisture can have a significant influence  
6 on precipitation. Only recently however have there been attempts to quantify this coupling, from a statistical  
7 viewpoint (Dirmeyer, 2001; Koster et al., 2001, 2002; Reale and Dirmeyer, 2002; Reale et al., 2002; Koster  
8 et al., 2003. Koster and Suarez (2004) analyse statistically records of precipitation over the US and find  
9 positive “memory” in the data for at least 3 months into the future. As previously mentioned, Koster et al  
10 (2004) report on a new model intercomparison activity, the Global Land Atmosphere Coupling Experiment  
11 (GLACE), that compares differences in the variability of precipitation among a large number of major  
12 climate models that is caused by interaction with soil moisture. They do this by an experimental protocol  
13 where they generate ensembles of simulations with and without soil moisture prescribed from a single  
14 simulation. They report a wide range of differences between models - apparently we can yet have no  
15 confidence in this important feedback component of a climate model and therefore its possible contribution  
16 to global warming simulations.

### 17 18 7.2.5.3 *Improved global and regional data*

19 Specification of land surface properties in climate models has been increasingly improved through new more  
20 accurate global satellite observations. They have provided in particular albedos of soils in non-vegetated  
21 regions (e.g., Tsvetsinskaya et al., 2002), (Wang et al., 2004), and emissivities (Zhou et al., 2003a,b), and  
22 constrain model calculated albedos in the presence of vegetation (Zhou et al., 2003d; Oleson et al., 2003),  
23 vegetation underlain by snow (Jin et al., 2002), and the role of leaf area (Tian et al., 2004a-c). Radiative  
24 temperatures are a key constraint (recent analyses include Oku and Ishikawa, 2004). Precipitation data sets  
25 combining rain gauge and satellite (Clem et al., 2002; Adler et al., 2003) are providing important diagnostic  
26 constraints for climate modelling, as are observations of runoff (Dai and Trenberth, 2002; Fekete, 2002).

### 27 28 7.2.5.4 *Field observational programs*

29 New and improved local site observational constraints collectively describe the land processes that need to  
30 be modelled. The largest recent such activity has been the LBA project in the Amazon (Malki et al., 2002).  
31 LBA studies have included physical climate on all scales, carbon dynamics, nutrient dynamics, and trace gas  
32 fluxes. The first of these is reviewed here. Goncalves et al. (2005) have discussed the importance of  
33 incorporating land cover heterogeneity in weather prediction models for South America. Rocha et al. (2005)  
34 and Quesada et al. (2005) have quantified water and energy budgets for a forested and a savanna site  
35 respectively. They find large differences in the water budgets between the *campo sujo* savanna at the IBGE  
36 Reserve and the dense forest at the Tapajós National Forest. Dry season evapotranspiration for the savannah  
37 averaged 1.6 mm day<sup>-1</sup> versus 4.9 mm day<sup>-1</sup> for the forest. Both ecosystems depend on deep rooting to  
38 sustain evapotranspiration during the dry season. Rocha et al. (2005) also observed that hydraulic lift  
39 recharged the forest upper soil profiles each night. At Tapajós, the forest showed no signs of drought stress  
40 allowing uniformly high carbon uptake throughout the dry season (July – December 2000) (Rocha et  
41 al.2005, and Goulden et al., 2005).

42  
43 Tibet as another key region continues to be better characterized from observational studies (e.g., Gao et al.,  
44 2004). With its high elevation, hence low air densities, land drives a much higher boundary layer than  
45 elsewhere. However, the water vapour mixing ratio is found to strongly drop off within the boundary layer  
46 (Yang et al., 2004).

## 47 48 7.3 **The Carbon Cycle and the Climate System**

### 49 50 7.3.1 *Contemporary Carbon Budget*

#### 51 52 7.3.1.1 *Global budget of atmospheric CO<sub>2</sub>*

53 Concentrations of atmospheric CO<sub>2</sub> have increased each year since measurements started in 1959 (Figure  
54 7.3.1a; Keeling et al., 2004; Conway and Tans, 2004), roughly parallel to emission rates. The average CO<sub>2</sub>  
55 increase for 2000–2004, based on the CMDL network, is 1.88 ppm yr<sup>-1</sup> or 3.99 Pg-C yr<sup>-1</sup> (with an uncertainty  
56 of ±0.3% or 0.3 ppm for the total change over 5 years). The *airborne fraction* (the proportion of fossil fuel  
57 emissions appearing as annual increment) averaged 0.55 from 1959 to 2000 (Figure 7.3.1b). Year-to-year

1 fluctuations in the airborne fraction occur associated with ENSO events, volcanic eruptions, and other major  
2 climatic events (Bacastow et al., 1980; Lintner, 2002; Lucht et al., 2002; Reichenau and Esser, 2003;  
3 Rödenbeck et al., 2003; Angert et al., 2004, 2005). However there is remarkably little variation in the 5-year  
4 mean values (horizontal lines in Figure 7.3.1b), with the important exception of the early 1990s. The annual  
5 increase in 1998, 2.5 ppm, was the highest ever observed, although the airborne fraction that year (0.82) was  
6 no higher than peak values observed in prior decades. On average, the terrestrial biosphere and the oceans  
7 together have consistently removed ~45% of the CO<sub>2</sub> emitted from fossil sources since 1959.

8  
9 [INSERT FIGURE 7.3.1 HERE]

10  
11 The concentration of CO<sub>2</sub> is currently higher in the Northern Hemisphere than in the South. The excess,  
12  $\Delta\text{CO}_2^{\text{N-S}}$ , has increased with emission rates of fossil fuel by about 0.5 ppm (Pg-C yr<sup>-1</sup>)<sup>-1</sup> (Figure 7.3.1c),  
13 giving a clear signature of the global impact of fossil fuel CO<sub>2</sub>. The intercept of the best-fit line is -0.8 ppm,  
14 indicating that, without anthropogenic emissions, CO<sub>2</sub> would be slightly higher in the Southern Hemisphere  
15 than in the North, presumably due to transport of CO<sub>2</sub> by the ocean circulation. The relationship between  
16  $\Delta\text{CO}_2^{\text{N-S}}$  and fossil fuel emissions has been generally consistent over the 45 years of record, but with inter-  
17 annual fluctuations as large as  $\pm 0.4$  ppm (Figure 7.3.1d). At least some of this variability may be attributed  
18 to changes in atmospheric circulation (Dargaville et al., 2003).

### 19 7.3.1.2 Carbon cycle variability from 1990 to 2003

20 The early 1990s featured anomalously strong global sinks for atmospheric CO<sub>2</sub>. The airborne fraction  
21 dropped to 0.4, and values of  $\Delta\text{CO}_2^{\text{N-S}}$  were rather low (Fig. 7.3.1). Manning and Keeling, (2005; Figure  
22 7.3.2) observed low rates of decline of atmospheric O<sub>2</sub>, and Miller et al. (2005) report a comparable shift in  
23 values for  $\delta^{13}\text{CO}_2$ , suggesting that uptake of atmospheric CO<sub>2</sub> by terrestrial ecosystems was largely  
24 responsible for increased efficiency of removing fossil fuel CO<sub>2</sub> from the atmosphere.

25  
26  
27 [INSERT FIGURE 7.3.2 HERE]

28  
29 Enhanced uptake of atmospheric CO<sub>2</sub> in the early 1990s has been attributed to the combined effects of the  
30 Mt. Pinatubo eruption and ENSO events (Reichenau and Esser, 2003; Rödenbeck et al., 2003), raising  
31 speculation that that negative feedback on rising CO<sub>2</sub> might lower projections of future increases. During  
32 this period, aerosols from the eruption of Mt. Pinatubo scattered sunlight and lowered atmospheric  
33 temperatures; there was a strong El Niño event, and the greenness index (NDVI) may have been lower  
34 globally. Lower temperatures are expected to reduce rates for respiration of labile pools in terrestrial  
35 ecosystems, and the ENSO may have simultaneously induced additional uptake of CO<sub>2</sub> by the ocean (Angert  
36 et al., 2005). Diffuse sunlight is used more efficiently than direct light by terrestrial ecosystems (Gu et al.,  
37 2003), offsetting reduced solar fluxes from the haze layer. Thus many influences of the eruption appeared to  
38 enhance uptake of CO<sub>2</sub> by terrestrial ecosystems. However, similar effects were not observed after the  
39 eruption of El Chichón, which was almost as large as the Mt. Pinatubo eruption and which also was  
40 associated with an El Niño event. Hence the explanation for this huge anomaly remains speculative.

41  
42 After 1995 the airborne fraction returned to values consistent with previous decades (Figure 7.3.1b). The  
43 decadal analysis of Manning and Keeling (2005) also indicates lower uptake by the terrestrial biosphere in  
44 1993–2003, as compared with 1990–2000 (Table 7.3.1). The remarkably high CO<sub>2</sub> growth in 1998 coincided  
45 with a global increase in CO concentrations attributable to wildfires. Achard et al. (2004) estimated  $0.88 \pm$   
46  $0.07$  Pg-C emitted from the burning of  $2.4 \times 10^6$  ha of peatland in the Indonesian forest fires in 1997–1998  
47 (Page et al., 2002), which would account for 0.4 ppm of CO<sub>2</sub> added to the atmosphere. There were also  
48 unusually large forest fires in the boreal zone that year (Yurganov et al., 2004). From 1998–2003, extensive  
49 droughts in mid-latitudes of the northern hemisphere (Hoerling and Kumar, 2003) may have led to decreased  
50 photosynthesis and carbon uptake (Lotch et al., 2005; Angert et al., 2005; Ciais et al., 2005), helping to push  
51 up the airborne fraction towards historical values.

52  
53 **Table 7.3.1.** Decadal Global CO<sub>2</sub> budgets. The **bold** numerals show the **total amounts in Pg-C** for each  
54 decade. The *italics* give the *mean annual flux* for emissions and sinks, and the *change in ppm* as derived  
55 from atmospheric CO<sub>2</sub> and O<sub>2</sub> measurements (Manning and Keeling, 2005; also see Figure 2.2).

Fossil Emissions	Land Sink	Ocean Sink	Atmos. Rise	Time Period
<b>63.5</b> (3.00)	<b>14.2</b> (0.67)	<b>17.1</b> (0.81)	<b>32.2</b> (1.52)	01/01/1990 to 01/01/2000
<b>64.8</b> (3.05)	<b>5.1</b> (0.24)	<b>22.4</b> (1.05)	<b>37.3</b> (1.76)	01/01/1993 to 01/01/2003

We cannot infer trends in net uptake by land or ocean ecosystems over periods shorter than 5–10 years. Inter-annual variations in the ventilation of the ocean mixed layer (Manning and Keeling, 2005) can produce significant changes of the O<sub>2</sub>:N<sub>2</sub> ratio on a yearly basis. The N-S gradient in CO<sub>2</sub> is influenced by changes in atmospheric circulation, changes in rates of fossil fuel emissions and in the time derivative of emissions (due to the lag inherent in inter-hemispheric exchange) (Rödenbeck et al., 2003; Dragaville et al., 2003). The long-term stability of uptake rates suggests the absence of vast changes in the carbon cycle globally over the last 45 years, but unfortunately the diverse sources of variability can mask quite significant trends. Hence consistent, very carefully calibrated studies of atmospheric CO<sub>2</sub> are needed over long time periods to determine if, as suggested by data from recent year, CO<sub>2</sub> growth rates and/or airborne fractions are increasing, or if there has been a shift in the land/ocean balance.

### 7.3.2 Carbon Cycle Processes

#### 7.3.2.0 Understanding the global CO<sub>2</sub> budget

On average, 30–50% of the net removal of CO<sub>2</sub> from the atmosphere, and a larger fraction of the inter-annual variation, may be attributed to uptake by the terrestrial biosphere, and the balance is taken up by the oceans (e.g., Bousquet et al., 2000; Battle et al., 2000; Manning and Keeling, 2005; Table 7.3.1). The loci of uptake and release, and the underlying processes, must be understood if we are to predict the future trends of CO<sub>2</sub> concentrations in atmosphere. We summarize below current knowledge of the processes that regulate the carbon cycle on land (7.3.2.1) and in the oceans (7.3.2.2). We then summarize large-scale net CO<sub>2</sub> exchange and associated spatiotemporal variations inferred from data using bottom-up and top-down (inverse) models (7.3.3). Finally we examine the results of initial model studies that attempt to simulate interactions between climate change, large scale carbon fluxes, and atmosphere CO<sub>2</sub> concentrations, and we place these studies in context (7.3.4).

#### 7.3.2.1 Terrestrial vegetation and the global CO<sub>2</sub> budget

Factors affecting the carbon cycle in terrestrial ecosystems include: (1) *direct climate effects* (changes in precipitation, temperature, aerosol scattering), (2) *intrinsic ecosystem effects* (land use including deforestation, reforestation, afforestation and historical legacies), and (3) *atmospheric composition effects* (fertilization by rising CO<sub>2</sub> or inputs of nutrients, damage by pollution).

##### 7.3.2.1.1 Where is C being stored, where emitted, in the terrestrial biosphere

Carbon uptake and storage in the terrestrial biosphere is the net sum of uptake due to growth, reforestation, and sequestration and emission due to deforestation, forest damage by pollution, and disturbance of soils. Forests (live and dead trees, soils) and peatlands contain the largest stocks of organic matter globally, and have the greatest potential to influence the global carbon cycle. Battle et al. (2001) argued, on the basis of changes in <sup>13</sup>CO<sub>2</sub>:<sup>12</sup>CO<sub>2</sub> and O<sub>2</sub>:N<sub>2</sub> that forest re-growth at middle latitudes roughly balanced deforestation in the tropics during the 1980s. Then, in the early 1990s, uptake of CO<sub>2</sub> and storage as organic matter by the terrestrial biosphere accelerated, and sinks exceeded sources due to deforestation since that time. Increasing forest uptake after 1990 could reflect slower rates of deforestation, or greater uptake by forests (see below), stimulated by changes in atmospheric composition, climate, and/or historical (land use) factors. The apparent return to values typical of earlier decades (noted above) suggests a transient global phenomenon rather than some kind of long-term stimulation (“greening”, cf. Idso et al., 1994) of the biosphere.

##### 7.3.2.1.2 Effects of land use change and exploitation of forest resources on global CO<sub>2</sub>

*CO<sub>2</sub> sources from tropical deforestation:* During the past 25 years, large-scale clearing of forests has been essentially a tropical phenomenon. Agriculture and exploitation of forest resources have reached into formerly remote areas of old-growth forest in the tropics, in contrast to mid-latitudes where exploitation previously eliminated old-growth. DeFries et al. (2002a) and Achard et al (2004) estimated net release of CO<sub>2</sub> from tropical deforestation and regrowth in the range 0.6 (0.3–0.8) and 0.9 (0.5–1.4) Pg-C yr<sup>-1</sup> for the 1980s and 1990s, respectively (DeFries), and 0.98 ± 30 Pg-C yr<sup>-1</sup> in the 1990s (Achard), using satellite data and terrestrial carbon models (see Table 7.3.2). Changes in forest area were found to be lower than given by

the United Nations Food and Agriculture Organization Forest Resource Assessment (FRA), 1.9 (0.6–2.5) Pg-C yr<sup>-1</sup> (Houghton et al., 2003). The Defries and Achard estimates are broadly consistent with the global budget for terrestrial systems, but it is harder to accommodate the large source estimated by Houghton.

**Table 7.3.2.** Estimated carbon emissions from tropical deforestation in the 1990s (Pg-C yr<sup>-1</sup>).

Author	Tropical Americas	Tropical Africa	Tropical Asia	Pan-Tropical
Houghton	0.75 ± 0.3	0.35 ± 0.2	1.09 ± 0.5	2.20 ± 0.6
De Fries	0.43 (0.21–0.62)	0.12 (0.08–0.14)	0.35 (0.19–0.59)	0.91 (0.50–1.36)
Achard	0.44 (0.35–0.52)	0.16 (0.13–0.19)	0.39 (0.32–0.56)	0.98 (0.81–1.12)

Note:

Differences mainly reflect estimates of deforested areas, higher for Houghton (FAO deforestation statistics) than for de Fries et al and Achard et al. (independently analyzed satellite data with wall-to-wall coverage).

#### 7.3.2.1.3 Forest re-growth in middle and high latitudes

In contrast to the tropics, forest areas generally increased during the 20th century at middle and high latitudes. This surprising trend reflects the intensification of agriculture and forestry. More food is being grown on less land, reflecting mechanization of agriculture, increased fertilizer use, and adoption of high-yield cultivars. Likewise intensive forest management and agroforestry produce more fiber on less land; improved forest management favors more rapid regrowth of forests after harvest. These trends have clearly led to carbon sequestration by re-growing forests, with beneficial effects on global concentrations of CO<sub>2</sub>. It should be noted however that industrialized agriculture and forestry require high inputs of fossil energy, so it is difficult to assess the net global effects of agricultural intensification on the atmosphere.

Regional studies have confirmed the plausibility of strong mid-latitude sinks due to forest re-growth. Networks of eddy flux towers have been developed where the exchange fluxes of CO<sub>2</sub> are measured directly, every hour, for a decade or longer (Baldocchi et al., 2003). These observations show that forests on long-abandoned former agricultural lands (Curtis et al., 2002; Valentini et al., 2004) and in industrial managed forests (Hollinger et al., 2002) take up significant amounts of carbon every year. Analysis of forest inventory data shows that, in aggregate, current forest lands are significant sinks for atmospheric CO<sub>2</sub> (Pacala et al., 2001). There are very few old-growth forests at mid-latitudes (most are less than 70 years old), and these forests are accumulating biomass simply because of their ages and stages of succession. The uptake rates inferred from flux towers are generally consistent with those inferred from inverse studies (e.g., Hurtt et al., 2003). Stocks of soil carbon are also likely increasing due to replenishment of soil organic matter and necromass depleted during the agricultural phase, and changes in soil microclimate associated with reforestation; these effects might add 30–50% to the quantity of CO<sub>2</sub> sequestered in biomass (e.g., Barford et al., 2001).

The carbon balance of high latitude forests is less clear. Significant regrowth of forests appears to be taking place across Scandinavia and Russia (*citations*). But fire disturbance in North American and Eurasian boreal forests was higher in the 1980s than in any previous decade on record (Kurz and Apps, 1999; Kurz et al., 1995, *citation needed for Eurasia*), and significant soil organic matter appears to be released in these fires. Permafrost in many areas (e.g., Alaska: Osterkamp and Romanovsky, 1999) is also currently warming. These processes are both likely to lead to release of important quantities of CO<sub>2</sub> (Harden et al., 2000; Goulden et al., 1998). Increased fire frequency and thawing of frozen soils may both be linked to climatic variations and trends (Zuang et al., 2003; Hess et al., 2001; Flannigan et al., 2000; Kurz and Apps, 1999), which feature notable warming in the latter part of the 20th century (Osterkamp and Romanovsky, 1999).

#### 7.3.2.1.4 Effects of climate variations and trends and changes in atmospheric composition on forest uptake or emission of CO<sub>2</sub>

*A CO<sub>2</sub> sink in undisturbed tropical forests?* Despite expanding areas of deforestation and degradation, there are still large areas of tropical forests that are among the world's great wilderness areas, with fairly light human impact, especially in Amazônia. Old growth tropical forests contain huge stores of organic matter, and they account for a major fraction of global net primary productivity (40% of global biomass, >60% of global NPP, Brown and Lugo, 1982). Changes in the carbon balance of these regions could have significant effects on global CO<sub>2</sub>.

1  
2 The net carbon balance of an area of forest may be measured by net primary production (input of organic  
3 matter) and heterotrophic respiration (output), or alternatively, ecosystem photosynthesis (input) and  
4 respiration (output). In many ecosystems, fire must be added on the output side, and in managed systems,  
5 harvesting must be taken into account. On long time scales, input and output must approximately balance,  
6 and changes in inputs eventually produce corresponding changes in outputs. Variations of inputs of organic  
7 matter, or of effective rates of mineralization, can result in a net flux of carbon to or from the forest system if  
8 they occur on a timescale shorter than the carbon residence times, even where basic ecosystem function is  
9 unchanged. For example, variations in cloudiness, precipitation and temperature might result in significant  
10 interannual variations in forest carbon balance. Bigger perturbations, such as episodes of mortality, or land  
11 management practices such as suppression of fires, can result in net fluxes that persist for decades at  
12 landscape or even continental scale (Pacala et al., 2001; Koerner, 2004).

13  
14 Data do not yet exist to compile a systematic forest inventory and carbon budget across the whole landscape  
15 in tropical regions. Recent investigations of the carbon balance of tropical forests have focused on long-term  
16 plots in mature, undisturbed ecosystems. These might be expected, on average, to be in carbon balance, and  
17 imbalances could indicate ecological responses to global atmospheric change. Indeed, Phillips et al. (1998)  
18 and Baker et al. (2004) reported that their forest plots appeared to be accumulating carbon at a mean rate of  
19  $0.7 \pm 0.2 \text{ t-C ha}^{-1} \text{ yr}^{-1}$ , implying net carbon uptake into live biomass of  $0.6 \pm 0.3 \text{ Pg-C yr}^{-1}$ , although they  
20 noted large variability between plots, due to the natural dynamics of tree death and regrowth..

21  
22 The rise in mean atmospheric CO<sub>2</sub> from a preindustrial value of 280 ppm to more than 375 ppm (in 2004)  
23 might cause net uptake of CO<sub>2</sub> simply by stimulating plant photosynthesis, since the respiration response  
24 will lag behind. The CO<sub>2</sub> concentration of the atmosphere has increased on average by  $\sim 1.5 \text{ ppm yr}^{-1}$ , 0.4%  
25 of the background concentration. The incremental stimulation of photosynthesis each year (estimated from  
26 biochemical studies) might average about 0.25% (e.g. next year's photosynthesis should be 1.0025 times this  
27 year's) (Berry references). Using of  $\sim 10$  years for the mean turnover rate for organic matter in tropical  
28 forests (Taylor and Lloyd, 1996), one might expect a present imbalance between uptake of CO<sub>2</sub> and  
29 respiration of 2.5% ( $1.0025^{10}$ ), consistent with rate of live biomass increase ( $\sim 3\%$ ) inferred from plot studies  
30 by Phillips et al. (1998).

31  
32 Other factors affecting these forests could be the pan-tropical rise in air temperatures by 0.26°C per decade  
33 in recent decades (Malhi and Wright, 2004), which could enhance nutrient mineralization, but alternatively  
34 this warming might increase water stress and respiration rates. Forest response to CO<sub>2</sub> might not be limited  
35 by light or by CO<sub>2</sub>, as assumed above, but by other factors such as low nutrient availability (Lewis, Malhi  
36 and Phillips, 2004; Koerner, 2004; Chambers and Silver, 2004), architectural constraints on how much  
37 biomass a forest can hold, ecosystem level constraints such as light competition, or differential responses  
38 that might favour shorter lived trees or agents of disturbance (insects, lianas) (Koerner, 2004). Indeed, Baker  
39 et al. (2004) reported higher mortality rates and increased prevalence of lianas in their plots. If these changes  
40 in forest dynamism occurred in the last 10 years, it is possible that even the *sign* of the imbalance inferred  
41 from plot measurements could be incorrect. Necromass was not determined, and areas with recent increases  
42 in dynamism, rates of decay of dead wood more than offset the increases in live biomass that occurred in  
43 response to mortality events (release of the subcanopy) (Rice et al., 2004).

44  
45 Another outstanding uncertainty is any methodological bias associated with sub-sampling of a natural  
46 disturbance and recovery matrix of the forest (with mature forests undergoing occasional local disturbance  
47 (e.g. storm blowdowns) followed by sustained periods of recovery. Subsampling of this matrix may leads to  
48 an artefactual apparent increase in biomass, depending on the size-frequency distribution of disturbances,  
49 and the recovery time of forests from such disturbances (Koerner, 2004). By far the biggest biomass  
50 increases were estimated for floodplain forests, where forests pass through a successional sequence starting  
51 at river-deposited gravel; the plots on these floodplains could involve forests in mid-succession, and thus be  
52 unrepresentative of the large-scale forest.

53  
54 In this context, it is noteworthy that studies involving large-area plots (9–50 ha) have indicated either no net  
55 long-term change or a long-term net decline in estimated above ground live biomass [1. *BDFFP site,*  
56 *Manaus, Brazil*, 18 1-ha plots in the PDBFF site, monitored 15 yr, Laurance et al., 2004; 2. *Barro Colorado*  
57 *I., Panama*, 50 ha plot, 15 yr, Chave et al., 2003; 3. *La Selva, Costa Rica*, 18 0.5-ha plots, 4 (-6) yr, Clark

1 2004a; 4. *JACARANDA* plots, Manaus, Brazil, two long-transect 5-ha plots, 6 yr, biomass data in Baker et  
2 al., 2004), and the 20 ha plot in Tapajós (5 years) showed increasing live biomass offset by decaying  
3 necromass (Saleska et al., 2003; Fearnside, 2000)]. Indeed, Körner (2004) argued that accurate assessment of  
4 trends in forest carbon balance requires long-term monitoring of many replicate plots or very large plots at  
5 each site in order to capture the localized and/or sporadic biomass changes associated with the natural forest  
6 disturbance regime.

7  
8 One of the key discoveries from the plot network has been that western Amazonian forests are 2–3 times  
9 more productive than eastern forests (Malhi et al., 2004), but similar or lower in biomass. Hence the  
10 residence times for live biomass carbon in western Amazonian forests is less than in eastern forests. This  
11 finding has implications for wider considerations of ecosystem response to stimulations of productivity  
12 (whether by CO<sub>2</sub> or N deposition or anything else), suggesting the long-term response to increased  
13 productivity in a mature forest ecosystem may not be a large increase in biomass, but an increase in turnover  
14 rate. Dynamism of tropical forests may be much more responsive than biomass, and system-level response  
15 (decrease in lifetime) is very different from the plant-level response (increase in productivity) (Koerner,  
16 2004).

17  
18 If the results from the plots could in fact be extrapolated, the mean above ground carbon sink would be  $0.61$   
19  $\pm 0.22$  Pg-C ha<sup>-1</sup> yr<sup>-1</sup> (Baker et al., 2004), or  $0.89 \pm 0.32$  Mg-C ha<sup>-1</sup> yr<sup>-1</sup> including corrections for small trees,  
20 lianas, and below-ground biomass. Multiplying by the FAO estimate of Neotropical moist forest area  
21 (5,987,000 km<sup>2</sup>) gives a Neotropical moist forest biomass sink of  $0.54 \pm 0.19$  Pg-C yr<sup>-1</sup> (Malhi and Phillips  
22 2004) Scaling-up taking into account soil type would increase this value to about  $0.62 \pm 0.18$  Pg-C yr<sup>-1</sup>.  
23 Finally, if the as yet uncompiled data from the African and Asia tropics (which account for 50% of global  
24 moist forest area) were to show a similar trend to Amazonia, the global moist tropical live biomass sink  
25 would be about  $1.2 \pm 0.4$  Pg-C yr<sup>-1</sup>. This value is close to the net source inferred by DeFries and Achard  
26 (Table 7.3.2). Taken at face value, the combined net exchange of CO<sub>2</sub> between the atmosphere and tropical  
27 land ecosystems would be ~0, which is intriguingly consistent with inferences from inverse studies of little  
28 net flux from low latitudes.

29  
30 *Atmospheric composition effects:* Free Air CO<sub>2</sub> Enrichment (FACE) studies have been used to examine the  
31 response of ecosystems to a large (usually about 50%) step increase in CO<sub>2</sub> concentration. The results of  
32 these have been quite variable (Nowak et al., 2004). In some instances, the expected stimulation was not  
33 observed or decreased to near zero after a few years (Oren et al., 2001) or there was even an inhibition of  
34 growth by elevated CO<sub>2</sub> (Shaw et al., 2002). Nevertheless, on average some stimulation has been observed in  
35 FACE experiments—but not as much as predicted from the kinetics of photosynthesis. These results clearly  
36 demonstrate other factors (nutrients for example) can limit plant growth, and throttle the CO<sub>2</sub> fertilization  
37 effect. However, the CO<sub>2</sub> perturbation used in FACE experiments is roughly 100 fold larger than the annual  
38 rate of CO<sub>2</sub> increase imposed annually in nature. Therefore, one might expect stronger constraints on the  
39 CO<sub>2</sub> fertilization effect in the FACE experiments than might occur in nature. Overall, we do not presently  
40 have an accurate measure of the magnitude of the CO<sub>2</sub> fertilization effect, and it may be quite difficult to  
41 determine by direct measurement due to the strong influence of other factors such as succession, disturbance,  
42 etc.

43  
44 Nutrient and pollutant deposition are considered in sections 7.4.2 and 7.5.1.

#### 45 46 7.3.2.1.7 *Direct effects of land cover on climate*

47 Land cover demonstrably affects both surface and regional climate. In the past several centuries, large-scale  
48 land-cover change occurred primarily in temperate latitudes through conversion of forests and grassland to  
49 agriculture; and then reversion to forests. Both the surface and regional climate, and global energy balance,  
50 have been affected. The dominant effect of forest regrowth in midlatitudes on the surface climate record has  
51 likely been cooling during summer, due to increased conversion of incoming radiation to latent heat and  
52 increases in boundary-layer cloudiness (DeFries et al., 2002b; Freedman et al., 2001); the effect was cooling  
53 0.3–0.6 C between 1910 and 1990, according to Roy et al. (2003). A particularly strong effect is noted in late  
54 spring when leaves emerge on trees (Collatz et al., 2000; Durre and Wallace, 2001; Bonfils et al., 2004,  
55 2005), with long-term trends associated with replacement of sparsely-vegetated croplands by forests  
56 (Fitzjarrald et al., 2001). Modelling studies (e.g., Snyder et al., 2004; Oyama and Nobre, 2004) and analysis  
57 of historical data appear to agree on these effects. Trends have likely been the opposite in the tropics due to

1 deforestation there. Freedman et al. (2001) pointed out that there is a feedback between forest cover and CO<sub>2</sub>  
2 uptake. Analyzing data for New England, they showed that forest cover increases boundary-layer cloudiness  
3 in late spring and early summer, and that CO<sub>2</sub> uptake was enhanced by the resulting increase in diffuse light.  
4

5 More uncertain is the influence of land cover on global climate. DeFries et al. (2002b) argued that future  
6 deforestation would warm the near-surface atmosphere by up to 2°C and lead to drier hydrologic conditions  
7 in the tropics. Warming appears likely over reforested land areas due to lower albedo, and cooling over  
8 deforested land areas (Brovkin et al., 1999, 2004). Overall, global effects appear to be rather small.  
9 Nevertheless the influence of land cover change on climate *data*, and thus on trends inferred from those data,  
10 may be more significant. Thus the most likely effects of changes in forest cover on climate have been to  
11 mask regional warming trends at surface stations in midlatitudes, and enhance warming trends in the tropics.  
12

### 13 7.3.2.2 *Ocean carbon-cycle processes*

14 So far, the oceans have taken up 48% of the anthropogenic CO<sub>2</sub> emissions from fossil fuel burning and  
15 cement manufacturing (Sabine et al., 2994). Changes in climate will affect the ocean carbon cycle (for  
16 observations, see Chapter 5, Sections 5.4.2 and 5.4.3), which itself will feed back onto climate change. So  
17 far, a series of marine carbon cycle climate feedbacks were identified. The as yet missing explanation of the  
18 glacial-interglacial CO<sub>2</sub> drawdown in the atmosphere indicates a gap in our knowledge with respect to  
19 marine processes, which are significant for biogeochemical forcing of climate (see paleo-climate carbon Box  
20 6.2 in Chapter 6). It is possible that a series of smaller feedbacks add up to an overall important feedback.  
21 Future impacts of the marine carbon cycle onto the climate system are not only triggered by climatic forcing  
22 alone, but also depend on further other anthropogenic forcings such as land use, water treatment, and  
23 building of freshwater reservoirs. One of the most important forcings, namely the emissions of fossil fuel  
24 CO<sub>2</sub>, depends on human behaviour and not on climate itself, although climate change may influence human  
25 behaviour. Moreover, marine carbon cycle feedbacks work on a variety of time scales. Long-time scale  
26 processes would persist for several tens of thousands of years, even when anthropogenic CO<sub>2</sub> emissions  
27 would stop immediately. Marine biogeochemical feedbacks of climatic relevance are summarised in Figure  
28 7.3.3 and Table 7.3.3.  
29

30 [INSERT FIGURE 7.3.3 HERE]

31 [INSERT TABLE 7.3.3 HERE]

32  
33  
34 In Table 7.3.3, we discriminate the feedbacks by their respective original forcing. Some of the feedbacks  
35 have wider implications than just a direct influence on the climate system. These implications are mentioned,  
36 if they seem to be of relevance for the climate change context. Table 7.3.3 reflects that only a few ocean  
37 climate biogeochemical feedbacks are well established. These are the feedbacks involving the inorganic  
38 carbon cycle. Organic carbon cycle feedbacks – including destabilisation of gas hydrates and purposeful CO<sub>2</sub>  
39 storage – are not well known due to the extreme complexity of processes involved and the difficulty of  
40 observing these processes. This does not mean that the organic carbon cycle may be not important for  
41 climate. We simply do not know at present. The basics of marine carbon chemistry and ocean acidification  
42 are explained in Box 7.1.  
43

44 [START OF BOX 7.1]

#### 45 **Box 7.1: Marine Carbon Chemistry and Ocean Acidification**

46  
47  
48 In the atmosphere, CO<sub>2</sub> is fairly inert and stays there mainly as molecular CO<sub>2</sub>. In contrast, any CO<sub>2</sub> which  
49 enters the ocean through air-sea gas exchange is hydrated to carbonic acid H<sub>2</sub>CO<sub>3</sub> and dissociated into “free  
50 CO<sub>2</sub>” (CO<sub>2</sub> plus H<sub>2</sub>CO<sub>3</sub>), HCO<sub>3</sub><sup>-</sup> (bicarbonate), CO<sub>3</sub><sup>2-</sup> (carbonate), and protons H<sup>+</sup>. The ratio of the respective  
51 concentrations [CO<sub>2</sub> plus H<sub>2</sub>CO<sub>3</sub>], [HCO<sub>3</sub><sup>-</sup>] and [CO<sub>3</sub><sup>2-</sup>] is roughly 1:100:10 but varies considerably. Their  
52 sum is called total dissolved inorganic carbon, [TCO<sub>2</sub>]. The ability of seawater to dissociate carbonic acid is  
53 quantified through the variable “alkalinity” (TAlk) which couples the marine carbon cycle to all other  
54 chemically active substances in seawater including water itself. The alkalinity is mainly determined by the  
55 cycles of calcium carbonate and borate. Thus the alkalinity inventory of the oceans as a whole varies usually  
56 on geological time scales only. In equilibrium, the CO<sub>2</sub> partial pressures (pCO<sub>2</sub>) in the atmosphere and the  
57 underlying seawater are equal. The higher this equilibrium CO<sub>2</sub> partial pressure is, the more carbonic acid is

1 dissociated in the ocean, and the higher is the proton concentration. A higher proton concentration means  
2 more acid conditions, i.e., a decrease in pH value ( $\text{pH} = -\log_{10}([\text{H}^+])$ ). In parallel to increasing seawater  $\text{pCO}_2$ ,  
3 the concentrations  $[\text{CO}_2 \text{ plus } \text{H}_2\text{CO}_3]$ ,  $[\text{HCO}_3^-]$ , and  $[\text{H}^+]$  increase while  $[\text{CO}_3^{2-}]$  decreases. The relative  
4 amount of  $\text{CO}_2$  additions to the ocean, which can be buffered through dissociation of carbonic acid,  
5 decreases with rising  $\text{pCO}_2$ : the “buffer factor” (see Figure 7.3.5) decreases, i.e. the seawater becomes less  
6 efficient for additional further  $\text{CO}_2$  uptake. At a given equilibrium  $\text{pCO}_2$ , alkalinity, temperature, and salinity,  
7 the concentrations  $[\text{HCO}_3^-]$ ,  $[\text{CO}_3^{2-}]$ , and  $[\text{H}^+]$  are uniquely determined (see Box 7.1, Figure 1) through the  
8 mass action law, and one cannot change one component without adjusting also the others.  
9

10 [INSERT BOX 7.1, FIGURE 1 HERE]  
11

12 The ocean acidification through uptake of anthropogenic  $\text{CO}_2$  leads to more corrosive conditions for calcium  
13 carbonate ( $\text{CaCO}_3$ ) in the ocean (“coffee machine analog”), i.e. towards an increase of areas which are  
14 undersaturated with respect to  $\text{CaCO}_3$ . The dissolution or preservation of  $\text{CaCO}_3$  in seawater is set by the  
15 solubility product:  $K_{\text{sp}} = [\text{Ca}^{2+}] \times [\text{CO}_3^{2-}]$ . At lower  $[\text{CO}_3^{2-}]$  in the water column during increasing  
16 acidification, two primary effects are expected: (1) the dissolution of  $\text{CaCO}_3$  at the ocean floor (available  
17 corals, available  $\text{CaCO}_3$  sediments) will be increasingly furthered, and (2) the biocalcification within the  
18 water column may be inhibited or slowed down (less biological production of corals as well as calcifying  
19 phytoplankton and zooplankton) (Royal Society, 2005). The metastable  $\text{CaCO}_3$  form aragonite (corals,  
20 pteropods) will be particularly susceptible to a pH lowering. The dissolution of  $\text{CaCO}_3$  sediments will occur  
21 on a very long time scale involving ten thousands of years and slowly increasing the alkalinity including the  
22 concentration of the carbonate ion  $\text{CO}_3^{2-}$ . On very long time scales this alkalinity compensation will be  
23 quantitatively significant for the neutralization of the anthropogenic  $\text{CO}_2$ . Or vice versa, the anthropogenic  
24  $\text{CO}_2$  invasion will impact on the deep sea  $\text{CaCO}_3$  sediment for tens of thousands of years, even if  
25 anthropogenic  $\text{CO}_2$  emissions would be halted immediately (Archer, 2005). For the carbon cycle, the  
26 hampering of biocalcification at the sea surface will probably not be quantitatively as important as the  
27  $\text{CaCO}_3$  dissolution on the sea floor but will occur immediately with decreasing surface ocean pH. A decrease  
28 in biocalcification alone will act as a small negative feedback (less alkalinity or  $\text{CO}_3^{2-}$  consumption) to  
29 atmospheric  $\text{CO}_2$ , but the effect of the marine particle flux (decreasing particle sinking velocities) may act as  
30 a positive feedback. Potential ecological changes due to ocean acidification may be severe, especially for  
31 corals in tropical, stably stratified waters, but also for cold water corals, and may influence the marine food  
32 chain up to higher trophical levels. These ecological changes therefore also have a considerable socio-  
33 economic impact. Apart from the absolute magnitude of a pH lowering, the expected fast rate of pH change  
34 at the sea surface can cause difficulties for marine ecosystems to adjust accordingly. Since the beginning of  
35 the industrial revolution, the sea surface pH dropped by about 0.1 pH units (corresponding to a 30% increase  
36 of the hydrogen ion concentration). Assuming a realistic emission scenario for anthropogenic  $\text{CO}_2$  such as  
37 the A1B scenario, biocalcification will become difficult in particular within the Southern Ocean realm by  
38 year 2100. Further details are found in Royal Society (2005). It is important to state that ocean acidification  
39 is not per se a consequence of climate change but a consequence of fossil fuel  $\text{CO}_2$  emissions, which are  
40 themselves the main driver of the anticipated climate change. Therefore, the issue needs to be addressed by  
41 both climate change communities and global change scientists in a broader approach.  
42

43 [END OF BOX 7.1]  
44

#### 45 7.3.2.2.1 *Warming of the ocean surface water*

46 The warming of the surface ocean as expected under a further accumulation of greenhouse gases in the  
47 troposphere has effects on the inorganic as well as the organic carbon cycles.  
48

49 The solubility of  $\text{CO}_2$  gas in seawater as well as the two dissociation constants of carbon acid in seawater  
50 depend on temperature and salinity (and also pressure) (Weiss, 1974; Millero et al., 2002). Warming of the  
51 water reduces the solubility and dissociation of  $\text{CO}_2$  and acts towards higher  $\text{pCO}_2$ . For a  $1^\circ\text{C}$  increase of the  
52 sea surface temperature, a change of 4.9–7.0  $\mu\text{atm}$  in atmospheric  $\text{pCO}_2$  results after 100–1000 yr taking into  
53 account the solubility effect only, and 6.9–10.2  $\mu\text{atm}$  for a combined effect of the temperature increase on  
54 solubility and dissociation constants after 100–1000 yr (use of the model of Heinze et al., 2003; see also  
55 Plattner et al., 2001; Broecker and Peng, 1986). The effect of rising seawater temperatures may be partially  
56 compensated by increasing fresh water releases to the ocean due to melting of ice sheets (see 7.3.2.2.2).  
57 Detailed changes in hydrographic conditions in the ocean surface layer can lead to further adjustments of the

1 solubility feedback. The solubility and dissociation feedback on a warming climate is positive, potentially in  
2 the order of few tens of  $\mu\text{atm}$  in atmospheric  $\text{pCO}_2$  increase and associated with uncertainties due to the  
3 difficulty in to predicting hydrographic surface ocean conditions. The feedback starts to build up  
4 immediately but depends on the turnover time of the ocean surface waters and their overall warming.  
5

6 Photosynthesis in the surface ocean is by far the largest contribution to biological primary production in the  
7 world ocean. On a yearly average (averaged over one seasonal cycle), the export of biologically bound  
8 carbon out of the surface ocean is the critical variable for potential changes of the atmospheric  $\text{pCO}_2$  due to  
9 biological process. Species composition and rain ratio changes (rain ratio = ratio of C atoms which are fixed  
10 to particulate organic carbon POC to those which are fixed to particulate inorganic carbon PIC, namely  
11  $\text{CaCO}_3$ , by biota) are discussed under 7.3.2.2.3 and 7.3.2.2.7. As long as the ocean circulation and nutrient as  
12 well as carbon supply remain constant, without any further feedback, the biological cycling of carbon would  
13 have only a small influence on the atmospheric  $\text{pCO}_2$  due to the non-linearity of the carbon chemistry (see  
14 7.3.2.2.7). It is difficult to quantify the direct effect of temperature on the uptake kinetics of marine biota for  
15 carbon and nutrients. According to the model of Michaelis-Menten kinetics, the uptake velocity of marine  
16 phytoplankton for nutrients and carbon approaches a maximum value ( $V_{\text{max}}$ ) at sufficiently large nutrient  
17 availability and decreases at low nutrient availability. The indicator is the half saturation constant  $K_s$ , i.e. the  
18 value of the ambient nutrient concentration where the uptake velocity is  $V_{\text{max}}/2$ . Though it may be expected  
19 that higher temperatures favor an acceleration of chemical reactions there is no conclusive formulation for  
20  $V_{\text{max}}$  at hand. Behrenfeld and Falkowski (1997, see their Figure 4) summarise a set of available  $V_{\text{max}}(T)$   
21 relationships in the form of maximum chlorophyll specific carbon fixation rates from different models,  
22 which implies that a pure temperature dependency may be difficult to deduce. The sign and magnitude of the  
23 biological productivity feedback to warming of the surface water are not yet known. Given a strong increase  
24 in  $V_{\text{max}}$  with increasing surface water temperature, potentially a negative feedback with a change of several  
25 tens of atmospheric  $\text{pCO}_2$  is possible (e.g. Heinze et al., 1991).  
26

27 Dissolved organic carbon (DOC) is considered to be stored in the ocean as a refractory pool with a very low  
28 turnover rate (Bauer et al., 1992) and a more semi-labile fraction (Carlson et al., 1994; Børshheim and  
29 Mykkestad, 1997; Loh et al., 2004). The oceanic inventory of the semi-labile DOC fraction is estimated to be  
30 of about the same order as the atmospheric  $\text{CO}_2$  inventory (Mopper and Degens, 1979; Hedges, 1992). The  
31 composition of dissolved organic matter in the ocean is still widely unknown (Loh et al., 2004). Both DOC  
32 fractions may react to an increase in the ambient seawater temperature, e.g., due to increased bacterial  
33 activity at higher temperatures. This feedback is expected to be positive, i.e., enhancing a climatic warming.  
34

#### 35 7.3.2.2.2 *Freshening of the ocean surface*

36 A freshening of the seawater due to additional inputs from melt water (sea ice, ice sheets, shelf ice) under a  
37 warmer climate will lead on the average to a negative feedback concerning the atmospheric  $\text{pCO}_2$ . Of course,  
38 the net effect depends on the vertical redistribution of the respective salinity anomaly. The effect would be  
39 larger if a melt water lid would cover the ocean rather than the additional freshwater being well mixed with  
40 the seawater. For a 0.5 psu decrease of the sea surface salinity, a change of  $-0.48$  to  $-0.59 \mu\text{atm}$  in  
41 atmospheric  $\text{pCO}_2$  results after 100–1000 yr taking into account the solubility effect only, and  $-2.1$  to  $-3.1$   
42  $\mu\text{atm}$  for a combined effect of the temperature increase on solubility and dissociation constants after 100–  
43 1000 yr (use of the model of Heinze et al., 2003; see also Plattner et al., 2001; Broecker and Peng, 1986).  
44 The expected negative feedback thus is small.  
45

#### 46 7.3.2.2.3 *Rising $\text{CO}_2$ and acidification (pH lowering)*

47 The uptake of anthropogenic  $\text{CO}_2$  by the ocean leads to shift in the dissociation of free carbon dioxide to bi-  
48 carbonate and carbonate and induces a respective acidification or pH value decrease in seawater (see Box  
49 7.3, but also, e.g., Harvey, 1969; Caldeira and Wickett, 2003) (Figure 7.3.4). This shift in the inorganic  
50 carbon equilibrium has consequences for both the inorganic carbon cycle and biological feedback processes.  
51

52 [INSERT FIGURE 7.3.4 HERE]  
53

54 For a quantification of the uptake of anthropogenic of  $\text{CO}_2$  by the ocean two different issues must be  
55 considered: the kinetics of the uptake (how quick or slow is excess  $\text{CO}_2$  taken up from the atmosphere) and  
56 the ultimate uptake capacity (how much can be stored in the ocean at final equilibrium). For a quick look,  
57 one can consider an ocean surface layer of a given thickness and determine how much excess  $\text{CO}_2$  from the

1 atmosphere will be taken up by this layer. This approach has been pursued by Bolin and Eriksson (1959).  
2 The simultaneous change in pCO<sub>2</sub> (or [CO<sub>2</sub>], i.e., the CO<sub>2</sub> concentration in water, if the solubility is known)  
3 and the change in total dissolved inorganic carbon concentration [DIC] in a water parcel of arbitrary volume  
4 equilibrating with the atmosphere can be expressed as a ratio called buffer factor or Revelle factor (e.g.  
5 Zeebe and Wolf-Gladrow, 2001; after Roger Revelle 1909–1991, TA = const. indicates that the alkalinity of  
6 the water – i.e. its ability to dissociate acids such as H<sub>2</sub>CO<sub>3</sub> – is assumed to be constant):  
7

$$8 \quad \text{RF}_0 = \left\{ \frac{d[\text{CO}_2]/[\text{CO}_2]}{d[\text{DIC}]/[\text{DIC}]} \right\}_{\text{TA}=\text{const.}}$$

9

10 For a preindustrial pCO<sub>2</sub> value of 280 μatm this buffer factor amounts to about 8.4. This means that for a 1%  
11 change in the ocean storage of DIC the pCO<sub>2</sub> in the water changes by 8.4%. The buffer factor depends on  
12 temperature (see also 7.3.2.2.1), salinity (see also 7.3.2.2.2), and the pCO<sub>2</sub> (or pH value, as pH sinks with  
13 increasing pCO<sub>2</sub>). The buffer factor increases quasi-linearly with rising pCO<sub>2</sub> and reaches a value of about  
14 10.2 at 500 μatm pCO<sub>2</sub> (Figure 7.3.5). This means, that the increase in ultimate buffer capacity of a seawater  
15 parcel slows down as pCO<sub>2</sub> rises. This positive feedback of the inorganic oceanic carbon system on climate  
16 warming forcing is very well known. Details of its realisation depend on the kinetics for the CO<sub>2</sub> uptake,  
17 mainly on the renewal of surface waters by waters which have not yet equilibrated with the atmospheric  
18 CO<sub>2</sub>. Given that the present ocean circulation would remain constant with further rising atmospheric pCO<sub>2</sub>,  
19 the stably stratified mid and low latitude oceans will get increasingly less efficient as sinks for atmospheric  
20 CO<sub>2</sub> and the high latitude oceans – where deep water production occurs – will relatively gain in importance  
21 as “bottleneck” regions for downward CO<sub>2</sub> transport. The fact that the area of stably stratified ocean waters  
22 by far exceeds the hydrostatically less stable polar regions, and the anticipated slowing down of meridional  
23 overturning circulation, make this feedback quantitatively even more significant.  
24

25 [INSERT FIGURE 7.3.5 HERE]  
26

27 The impact of anthropogenic ocean acidification on the ocean saturation state with respect to CaCO<sub>3</sub> is  
28 already detectable (Feely et al., 2004; Orr et al., 2005). This acidification of seawater may have a significant  
29 impact on the biocalcification of marine organisms, corals and calcifying plankton organisms  
30 (coccolithophoridae, pteropods, foraminifera) in all surface ocean provinces, (Royal Society, 2005), as well  
31 as other bottom dwelling calcifiers in shallow waters (the effect on deep CaCO<sub>3</sub> sediments is discussed in a  
32 following paragraph). In laboratory experiments with the coccolithophore species *Emiliana huxleyi* and  
33 *Gephyrocapsa oceanica*, a significant reduction in CaCO<sub>3</sub> (PIC) production and a stimulation of particulate  
34 organic carbon (POC) production resulted under high ambient CO<sub>2</sub> partial pressure (Zondervan et al., 2001;  
35 Riebesell et al., 2000). Respective studies are being repeated at present, and there are indications that other  
36 species also may show different reactions, even with a reversal in the sign of the change (Tortell et al.,  
37 2002). Under nitrogen limiting conditions, even a decrease in both CaCO<sub>3</sub> production and POC production  
38 was detected in a laboratory experiment at high CO<sub>2</sub> partial pressures, so that no conclusive quantification of  
39 the CaCO<sub>3</sub> feedback is possible at this stage (Sciandra et al., 2003). The relative small negative feedback on  
40 a reduced CaCO<sub>3</sub> production on atmospheric pCO<sub>2</sub> may be compensated for by a change of the ballast for  
41 settling biogenic particles (Heinze, 2004; see also 7.3.2.2.7) and the associated shallowing of  
42 remineralization depth levels in the water column for organic carbon. The potential reduction in  
43 biocalcification provides probably only a minor feedback, but may have a major ecological impact, such as  
44 shifts in species composition. So far an increase of harmful algae blooming is observed, which so far,  
45 however, is mainly associated with a warming of the sea surface layer (Mudie et al., 2002; Van Dolah,  
46 2000). Rising pCO<sub>2</sub> leads also to a reduction of coral reefs, which are particularly susceptible to an  
47 acidification as they consist on the less stable CaCO<sub>3</sub> mineral aragonite (Kleypas et al., 1999; Hughes et al.,  
48 2003). Likewise, shallow benthic calcifying organisms such as benthic foraminifera would be affected by the  
49 lowering in ocean pH. The feedback on atmospheric CO<sub>2</sub> due to the CaCO<sub>3</sub> dissolution would be negative  
50 (because the alkalinity changes twice as much as the dissolved organic carbon concentration during CaCO<sub>3</sub>  
51 dissolution).  
52

53 While the corrosion of shallow water carbonate depositions and corals due to acidification occurs on a  
54 relatively short time scale, increased carbon storage in the ocean leads also to the dissolution of calcareous  
55 sediments in the deep sea (Broecker and Takahashi, 1978). This process is well known from sediment core  
56 analysis for the past climatic cycles (e.g., Balsam, 1983; Farrell and Prell, 1989). The feedback of CaCO<sub>3</sub>  
57 sediment dissolution on atmospheric pCO<sub>2</sub> is negative and quantitatively significant on a 10<sup>3</sup>–10<sup>5</sup> yr time

1 scale, where  $\text{CaCO}_3$  dissolution will account for a 60–70% compensation of the anthropogenic  $\text{CO}_2$   
2 emissions, while the ocean water column will account for 22–33% on a time scale of  $10^2$ – $10^3$  yr and the  
3 remaining 7–8% may be in addition compensated by long-term terrestrial weathering cycles involving  
4 silicate carbonates (Archer et al., 1998) (Figure 7.3.6). The ultimate extreme long-term overall buffering of  
5 the ocean including the  $\text{CaCO}_3$  sediment feedback amounts to approximately 11/12 of the atmospheric  
6 perturbation, leaving 1/11 airborne (Bolin and Eriksson, 1959). Due to the slow  $\text{CaCO}_3$  buffering  
7 mechanism, the mean atmospheric lifetime of anthropogenic  $\text{CO}_2$  is estimated to be 30,000-35,000 years  
8 (Archer, 2005).

9  
10 [INSERT FIGURE 7.3.6 HERE]

11  
12 A potential risk associated with the neutralization of anthropogenic  $\text{CO}_2$  through  $\text{CaCO}_3$  sediment  
13 dissolution is the destruction of wide areas of bottom and sediment fauna with so far not assessed impacts on  
14 the global biosphere, food chain, geochemical cycling and biodiversity. Potential feedbacks resulting from  
15 these potential changes are not known. Even with an immediate stop of excess  $\text{CO}_2$  emissions, the  
16 anthropogenic  $\text{CO}_2$  evasion to the atmosphere would leave its imprint on the marine sediment for the next  
17 several ten thousands of years.

18  
19 Elevated ambient  $\text{CO}_2$  levels appear also have an influence on the production rate of POC by marine  
20 calcifying plankton organisms (Zondervan et al., 2001). This increased carbon binding under higher  $\text{CO}_2$   
21 levels was also observed at three diatom species (diatoms are silicifying phytoplankton organisms)  
22 (Riebesell et al., 1993). For regulating the atmospheric  $\text{pCO}_2$  it is critical to know, whether these increased  
23 carbon fixation rates (which are also observed in terrestrial ecosystems) translate also into increased export  
24 production rates. This question so far could not be answered from laboratory experiments. Potentially  
25 dedicated in situ  $\text{CO}_2$  release experiments may address this question. Studies on the nutrient to carbon ratio  
26 in marine phytoplankton have not yet resulted in significant changes of the C:N:P ratio (“Redfield ratio”) in  
27 organic tissue (Burkhardt et al., 1999) with  $\text{CO}_2$  concentration. This insensitivity indicates that the  $\text{CO}_2$   
28 fertilisation effect may only be quantitatively significant in areas, where nutrients are not fully used up (such  
29 as the HNLC (high-nutrient-low-chlorophyll) areas of the Southern Ocean and few other selected regions).  
30 In these areas, however, other limiting factors may inhibit an increased POC production due to other factors  
31 such as lack of micronutrients (e.g., iron, zinc, Franck et al., 2003). Overall, a change of net carbon fixation  
32 with rising  $\text{pCO}_2$  is neither confirmed nor can it be ruled out. Magnitude and sign of the feedback under real  
33 world conditions are yet unknown (difficulties due to regional and species dependent conditions;  
34 experiments so far very selective; higher growth rates potentially could lead to more pulse like plankton  
35 blooms and a net increase in particle export from the euphotic zone).

36  
37 A lower pH of seawater influences all pH dependent chemical reactions where seawater is involved. Among  
38 others, the solubility of iron depends on pH. Measurements on Fe(III) hydroxides revealed a strong pH  
39 dependency between pH 2–7, but near constant solubilities in the pH range relevant for seawater (pH 7–8.5)  
40 (Byrne and Kester, 1976; Kuma et al., 1996; Liu and Millero, 2002). Therefore, a respective feedback of  
41 increased iron availability and stimulation of biological production under higher  $\text{CO}_2$  concentrations is  
42 unlikely to be significant.

#### 43 44 7.3.2.2.4 *Changes in cloud cover, sea ice cover, and incoming solar radiation*

45 Climate change will be associated with regional changes in cloud cover, sea ice cover, and hence incoming  
46 solar radiation into the surface ocean. Plankton production in the surface ocean depends on light availability  
47 and spectral composition (e.g., Jassby and Platt, 1976) and thus changes in the carbon fixation (changes in  
48 both the  $\text{CaCO}_3$  and POC production) are possible due to this shift in light availability. It is not known how  
49 the biologically controlled part of the carbon system in the polar oceans will change after a significant retreat  
50 of sea ice coverage. The absorption of incoming solar radiation by phytoplankton can regionally be  
51 important to feedback on ocean surface temperatures (Wetzel et al., under revision in 2005; Wetzel et al.,  
52 2004). Changes in incoming solar radiation will also be modulated by this feedback into potential  
53 temperature changes (see also 7.3.2.2.7). The climate feedback of this process is so far not accurately  
54 quantifiable.  
55

1 7.3.2.2.5 *Increasing stratification and reduction in large scale meridional overturning, shift of shelf*  
2 *regimes*

3 The ocean circulation is one of the “governing parameters” for the marine carbon cycle. Changes in the  
4 vertical overturning and stratification of the ocean change the supply for nutrients for biological carbon  
5 fixation, the alkalinity and dissolved inorganic carbon transport into the surface layer and away from it, and  
6 regulate the vertical redistribution of dissolved substances.

7  
8 In a simplified view one may assume that with a reduction of ocean circulation the rates of biological  
9 production may remain identical, because the slowing down of ocean circulation is compensated by a  
10 nutrient concentration increase (see Broecker and Peng, 1982; usually the nutrient concentration is an  
11 indicator for the age of open ocean waters). However, during the last glacial, a partial decoupling between  
12 shallow and deep large scale ocean circulation may have led to a vertical fractionation of nutrient as well as  
13 carbon concentrations and an efficient deep ocean storage of atmospherically derived carbon (Boyle, 1988).  
14 A respective storage of additional excess anthropogenic CO<sub>2</sub> in the deep-sea by a more sluggish ocean  
15 circulation under climatic warming may thus provide a negative feedback to atmospheric CO<sub>2</sub> on millennial  
16 time scales (reduced outgassing of already stored carbon). This negative feedback could be enhanced by an  
17 acceleration of CaCO<sub>3</sub> sediment dissolution in order to compensate for the excess CO<sub>2</sub>. In view of the  
18 different physical forcing conditions of the late glacial and future ocean circulations, an analog conclusion  
19 from the glacial ocean conditions may be misleading here (Crowley, 1990). The magnitude and sign of the  
20 feedback are not yet known for future conditions.

21  
22 A more sluggish ocean circulation and more pronounced density stratification would slow down the vertical  
23 transport of carbon and the replenishment of the ocean surface with water which has not yet been in contact  
24 with anthropogenic CO<sub>2</sub>. This narrowing of the “bottleneck” for anthropogenic CO<sub>2</sub> invasion into the ocean  
25 would provide kinetically a significant positive feedback on atmospheric greenhouse gas concentrations  
26 (also due to potential increases in N<sub>2</sub>O release from the ocean, see 7.3.2.2.7) (Bolin and Eriksson, 1959; see  
27 also the coupled climate model simulations by Cox et al., 2000; Friedlingstein et al., 2001). The efficiency of  
28 the buffer factor feedback (see 7.3.2.2.3) would be increased by the circulation feedback into a positive  
29 climate forcing feedback enhancing warming.

30  
31 Changes in ocean circulation in the course of a climatic shift can regionally affect shelf sea circulation  
32 systems. Different types of shelf sea systems have been identified (Walsh, 1991) that can result in either  
33 nutrients plus carbon export from the shallow seas into the open ocean or vice versa a lift of nutrients plus  
34 carbon onto the shelf and towards coastal areas. The North Sea may provide regionally a net sink for  
35 atmospheric CO<sub>2</sub> (Thomas et al., 2004), but this export will presumably be compensated by a net source in  
36 shelf sea systems of different type (Smith and Hollibaugh, 1993, i.e., the extrapolation to all shelf seas  
37 worldwide accounting for up to 20% of anthropogenic CO<sub>2</sub> uptake as made by Thomas et al., 2004, is not  
38 valid).

39  
40 In an equilibrium climate including steady state biogeochemical forcing, export and new production would  
41 be equal when globally integrated. During shifts of ocean circulation, transient mismatches may occur  
42 between global export production of biologically bound carbon plus carbon on one side and new production  
43 plus outgassing of carbon in upwelling areas respectively on the other side. Due to the regionally varying  
44 conditions and complexity of governing processes respective transient net CO<sub>2</sub> release or uptake events are  
45 likely to occur but are difficult to predict in detail.

46  
47 7.3.2.2.6 *Biogeochemical forcing (river loads, aeolian deposition, dust, micronutrients)*

48 Biogeochemical cycling in the ocean is ultimately fuelled by input of matter from the continents through  
49 aeolian deposition and river runoff loads (and to some degree also through ground water seeping and inputs  
50 from the earth’s interior). The ocean water column and top sediment act then as an interface between input of  
51 matter and output of matter back to the lithosphere.

52  
53 Input of carbon (DIC, DOC), alkalinity (ability of seawater to dissociate weak acids; approximated by [TA]  
54 = [HCO<sub>3</sub><sup>-</sup>] + 2[CO<sub>3</sub><sup>2-</sup>] + [B(OH)<sub>4</sub><sup>-</sup>] + [OH<sup>-</sup>] - [H<sup>+</sup>]), phosphate, nitrate (and ammonium), and silicic acid to  
55 the ocean takes place mainly through river runoff. The composition of river loads is both influenced by  
56 climate and anthropogenic activities. Rising CO<sub>2</sub> levels in the atmosphere and land use may lead to increased  
57 chemical and physical weathering resulting in increased carbon and alkalinity loads in rivers (for alkalinity

1 see Raymond and Cole, 2003; for dissolved organic carbon see Freeman et al., 2004; Hejzlar et al., 2003;  
2 Clair et al., 1999). Depending on the lithology and soil composition of the catchment areas, increased levels  
3 of alkalinity, DIC, or DOC can lead to local positive or negative feedbacks. Mobilisation of  $\text{CaCO}_3$  and  
4 silicate carbonates from soils and transfer to the ocean will lead to a negative feedback on atmospheric  $\text{CO}_2$   
5 (Dupre et al., 2003). Input of DOC and DIC from soils will lead to a positive feedback. Variations in nutrient  
6 supply, as induced by anthropogenic activities, can lead to local deviations from the large scale average  
7 P:N:C:Si ratios (“Redfield ratios”) in marine waters, especially near coasts, and change the biolimiting  
8 factors. Nutrient supply to the ocean has been changed through retention of silicic acid in freshwater systems  
9 due to the building of water reservoirs (e.g. Humborg et al., 2000) and increased nitrate release from land  
10 due to fertilizer use as well as nitrogen deposition from the atmosphere in highly polluted areas (Green et al.,  
11 2004; De Leeuw et al., 2001). These supply changes can lead to a species shift, biological production  
12 changes, anoxia, and methanogenesis in coastal waters. Nitrogen release through rivers has probably doubled  
13 from the beginning of the industrial revolution up to now (Green et al., 2004). Possibly the growth of  
14 calcifying organisms relative to silicifying plankton could be stimulated and result in a local positive  
15 feedback to atmospheric  $\text{CO}_2$ .

16  
17 Deposition of continentally derived matter from the atmosphere to the ocean surface provides an important  
18 source of micronutrients and ballast material to the ocean. Iron (also zinc and others, e.g., Frew et al., 2001)  
19 is a biolimiting micronutrient. Areas where iron is not supplied by aeolian transport in sufficient amounts  
20 tend to be iron limited resulting in elevated surface ocean concentrations of nutrients (HNLC regions). The  
21 aeolian supply of these micronutrients in a warmer climate depends on wind patterns, soil humidity, and  
22 lithology of land areas. A warmer climate may result on the average in a decrease of dust mobilisation and  
23 transport (Mahowald and Luo, 2003; Werner et al., 2002; though land use may result also in increased dust  
24 loads, Tegen et al., 2004). See paragraph 7.5.1.1 in this chapter for a detailed discussion on dust to  
25 climatically active aerosol. A decrease of dust loads would transfer less soluble or bioavailable iron to the  
26 oceans. It thus would lead to net positive feedback towards further increasing  $\text{CO}_2$  by a weakening of marine  
27 biological production. Reaction of dust particles with anthropogenic  $\text{SO}_2$  in the atmosphere, in contrast, may  
28 lead to a further mobilization of iron in mineral dust (Meshkhdze et al., 2003) and hence to a net fertilization  
29 of ocean waters, thus providing a negative climate feedback. Apart from chemical consequences of  
30 atmospheric dust deposition variations, a weakening of dust supply to the ocean surface can lead to a further  
31 positive feedback: Less clay ballast material may lead to smaller marine particle aggregates (biogenic matter  
32 produced within the ocean plus clay material brought in from the atmosphere) and to slower settling  
33 velocities of particles through the water column (Ittekkot, 1993; Haake and Ittekkot, 1990) (Figure 7.3.7).  
34 The consequence would be shallower remineralization depth levels and release of DIC higher up in the water  
35 column favouring increased outgassing of  $\text{CO}_2$  to the atmosphere (see also 7.3.2.2.7).

36  
37 [INSERT FIGURE 7.3.7 HERE]

#### 38 39 7.3.2.2.7 *Secondary feedbacks involving marine biological productivity (from changes in species* 40 *composition and particle flux mode, $\text{N}_2\text{O}$ cycle, DOC storage, TEP, DMS)*

41 Reactions of the marine carbon cycle to different climatic and anthropogenic boundary conditions can induce  
42 a series of further feedbacks to the climate system, which cannot straightforwardly be traced back to one  
43 single forcing. Quantifications of these feedback processes in an integrated way are still difficult.  
44 Nevertheless these feedback processes have to be addressed. Lack of detailed evidence does not rule out  
45 potential surprises within this complex system.

46  
47 The non-linearity of the marine carbon chemistry can provide a weak negative feedback in absolute terms of  
48 atmospheric  $\text{pCO}_2$  change. As the partial pressure in seawater increases non-linearly with increasing DIC,  
49 the same amount of carbon extracted from the surface ocean by marine biota will lead to a somewhat larger  
50  $\text{pCO}_2$  reduction at higher ambient  $\text{pCO}_2$ . Of course, remineralization of POC compensates for this reduction,  
51 so that this negative feedback would only be active at an increase of biogenic particle export production, and  
52 would turn into a positive feedback in case of a decrease in export production. The feedback would work  
53 instantaneously and is accurately quantifiable at the process level but difficult to quantify for the ocean as a  
54 whole.

55  
56 Changes in plankton species composition and regional shift of high production zones can lead to a series of  
57 further feedbacks. Light absorption due to these shifts by oceanic organisms may change and affect the

1 respective feedback on heating up the ocean surface water (Wetzel et al., *submitted in 2004*; Wetzel et al.,  
2 2004). An albedo change can be induced leading to either a negative or positive feedback to climate  
3 warming. The increased amount of blooms involving calcifying organisms as indicated for the high northern  
4 latitudes (Broerse et al., 2003; Smyth et al., 2004) can lead to a temporarily increased surface ocean albedo,  
5 though the effect on the radiation budget is quantitatively small (Tyrell et al., 1999). A positive or negative  
6 albedo feedback may also result from an areal reduction of coral reef systems or bleaching of corals, as the  
7 sediment composition in shallow water systems in principle influences the reflectance (Werdell and Roesler,  
8 2003) and hence the seawater temperature.

9  
10 Any ecological changes in the upper ocean may lead to a change in the vertical particle flux mode, in  
11 particular concerning the size as well as sinking speed of particle aggregates and the associated  
12 remineralization depth horizon. A deepening of this horizon would have the net effect of a surface ocean  
13 pCO<sub>2</sub> drawdown, a shallowing would result in an increase of surface ocean pCO<sub>2</sub> (see also 7.3.2.2.6 and  
14 7.3.2.2.3). There exists evidence for CaCO<sub>3</sub> to have an important ballast role in marine particle aggregates  
15 (Armstrong et al., 2002; Klaas and Archer, 2002). Therefore, any weakening of CaCO<sub>3</sub> export production  
16 may lead to a decrease in particle settling velocities potentially compensating (Heinze, 2004) or even  
17 overriding the negative feedback effect of increased alkalinity levels with less CaCO<sub>3</sub> production. Changes  
18 in the supply of iron for biological production can influence the thickness of biogenic silica shells of  
19 diatoms. Under lower iron availability, these opaline shells would be thicker and hence would increase  
20 particle settling velocities (Hutchins and Bruland, 1998). This negative feedback, however, may be  
21 overridden by the weaker biological production itself at iron stress plus the favouring of CaCO<sub>3</sub> shell  
22 material production if silicon is used up quicker as in a reference state. It is possible, that a surface nutrient  
23 depletion under global warming conditions leads to a weakening of biogenic silica production and a  
24 reduction of the ballast effect also due to less diatoms in marine particle rain. The corresponding favoring of  
25 small particles may lead to a positive feedback on atmospheric pCO<sub>2</sub> (Bopp et al., *submitted in 2005*). The  
26 marine inventory and distribution of DOC (dissolved organic carbon) may be susceptible to species shifts,  
27 warming, and CO<sub>2</sub> partial pressure, though no conclusive quantification of respective feedbacks can be  
28 provided at present (Engel et al., 2004). Potentially increased production of TEP (transparent exopolymer  
29 particles; Alldredge et al., 1993; Passow, 2002) during the build-up of biogenic particulate can accelerate the  
30 particle sinking through aggregate formation (Alldredge et al., 1993; Kiørboe et al., 1996; Passow et al.,  
31 2001).

32  
33 Ecological changes in conjunction with circulation changes can lead to either increases or decreases of low  
34 oxygen or even anoxic zones in the world ocean. Under oxygen depletion, denitrifying organisms can use the  
35 oxygen in nitrate for remineralization of organic matter. During the conversion of nitrate to molecular  
36 nitrogen, N<sub>2</sub>O also is produced which shows up as maxima below and above the O<sub>2</sub> minimum zones (Zehr  
37 and Ward, 2002). Outgassing of this N<sub>2</sub>O would contribute to a further increase of greenhouse gas  
38 concentrations in the atmosphere. Associated with anoxic marine conditions are further enhanced  
39 mobilizations of methane (Fuhrmann and Capone, 1991). On the other hand, nitrification at the sea surface  
40 can occur through biological N<sub>2</sub> fixation in coastal areas (Herbert, 1999) as well as in the open ocean (here in  
41 particular through the cyanobacterium *Trichodesmium*, Capone et al., 1997; Orcutt et al., 2001). N<sub>2</sub> fixation  
42 in principle would provide a means of strengthening biological carbon fixation; however, during N<sub>2</sub> fixation  
43 N<sub>2</sub>O also is produced which at least may partially compensate for such a negative feedback. For a lowering  
44 of pH values in seawaters, a reduction of N<sub>2</sub> fixation has been indicated (Huesemann et al., 2002). An  
45 increase of iron would stimulate N<sub>2</sub> fixation, so this process also depends on the iron supply (Raven and  
46 Falkowski, 1999) which may increase due to mobilisation in a more acid environment or decrease with less  
47 dust supply in a warmer and more humid climate.

48  
49 Dimethylsulphide (DMS) and its precursor DMSP are produced by a few classes of phytoplankton including  
50 specifically coccolithophores (CaCO<sub>3</sub> producing phytoplankton) (Matrai and Keller, 1993). DMS can be of  
51 climatic relevance as it can provide the basis for cloud condensation nuclei once it enters the atmosphere as  
52 marine aerosol (Shaw, 1983; Charlson et al., 1987; see also section 7.5.1.4 in this report. The “CLAW” (an  
53 acronym formed from the initials of the authors of Charlson et al., 1987) hypothesis suggests the following  
54 negative feedback loop: An increase in temperature and solar forcing would lead to an increase in biological  
55 production, which would induce a cooling effect due to production of DMS (and hence sulphate aerosol) and  
56 an associated cloud albedo (and possibly a reduction of atmospheric CO<sub>2</sub> through increased biological  
57 production in the ocean). So far, modelling studies on DMS production in a future warmer climate indicate

1 an increase of DMS production by 3% for a doubling (Bopp et al., 2004) and 14% for a tripling (Gabric et  
2 al., 2004) of the preindustrial pCO<sub>2</sub> in the atmosphere. The largest changes occur at high latitudes  
3 (specifically in the Southern Ocean) with only small changes in the tropics and sub-tropics. The expected  
4 negative feedback on radiative climate forcing is expected to be globally relatively small (−0.05 W m<sup>−2</sup> for  
5 2×CO<sub>2</sub>, Bopp et al., 2004), but may locally become significant.

#### 7.3.2.2.8 *Destabilization of methane gas hydrates*

8 Marine gas hydrates consist mostly of methane (CH<sub>4</sub>) and occur in sediment pore space under specific  
9 conditions of low temperatures, high pressure, and sufficient CH<sub>4</sub> availability, in particular at continental  
10 margins (Kvenvolden, 1993). If the state variables pressure and temperature are changed, CH<sub>4</sub> gas can  
11 potentially be set free in large quantities, an event which may have contributed to the latest Paleocene  
12 thermal maximum 55 million years before present (Dickens, 2001). The significance of gas hydrates for  
13 global change is not yet clarified (Kvenvolden, 2000, 2002). Potentially, larger amounts of CH<sub>4</sub> hydrate can  
14 be set free from the oceans also due to statically unstable continental margins and technical seabed  
15 installations (Beauchamp, 2004). It is possible that CH<sub>4</sub> from hydrates – once it is set free – is oxidized to  
16 CO<sub>2</sub> before entering the atmosphere (and thus have a less large impact than CH<sub>4</sub> itself on a further  
17 greenhouse gas increase) and may lead to massive microbial blooms in the world ocean (Valentine et al.,  
18 2001; Elvert et al., 2001). Potential destabilisation of gas hydrates would provide a positive feedback to  
19 global warming. Recently, the existing reservoir size CH<sub>4</sub> of gas hydrates was correct down with respect to  
20 previous estimates (ca. 500–2500 Pg-C) (Milkov, 2004). The potential use of these hydrates as a fossil fuel  
21 resource will have to be taken into account in emission scenarios.

#### 7.3.2.2.9 *Purposeful CO<sub>2</sub> storage in the ocean as an anthropogenic feedback to rising atmospheric pCO<sub>2</sub>*

24 Next to storage of anthropogenic CO<sub>2</sub> in terrestrial geological reservoirs, two options for purposeful oceanic  
25 CO<sub>2</sub> storage have been discussed: deep injection of CO<sub>2</sub> into the water column (e.g., Brewer et al., 2000) and  
26 large scale ocean surface fertilisation (e.g., Benemann, 1992). Purposeful storage of CO<sub>2</sub> would possibly  
27 provide a temporary negative feedback to climate change, while the long-term gross feedback would be zero  
28 or could potentially even turn into a positive one, e.g. if humans do not favour an energy saving strategy, but  
29 use the sequestration option to burn even more fossil fuel. Direct injection of anthropogenic CO<sub>2</sub> after its  
30 capture in the deep North Pacific Ocean appears to be the most efficient method (Orr, 2002), especially when  
31 the CO<sub>2</sub> is stored as liquid CO<sub>2</sub> lakes in isolated bathymetric features. The mitigation option of direct CO<sub>2</sub>  
32 injection into the water column is discussed in detail in the IPCC Special Report on Carbon Dioxide Storage  
33 and Capture, Chapter 6 (Caldeira et al., 2005.) The negative side effect on the local deep-sea ecosystems and  
34 unknown feedbacks of the bottom biogeochemical systems has to be taken into account. The impact of local  
35 changes of the chemical environment near CO<sub>2</sub> injection sites and liquid CO<sub>2</sub> pools over entire biomes and  
36 larger timescales is unclear (Caldeira et al., 2005). Fertilisation of the ocean in HNLC regions (especially the  
37 Southern Ocean) through addition of micronutrients, in particular iron, has been tested in several field  
38 experiments (e.g., Bakker et al., 2005; De Baar et al., 2005; Bishop et al., 2004). The poor efficiency of  
39 purposeful fertilisation has been documented over more than 10 years of research (De Baar, 1992; Watson et  
40 al., 1994; Cicerone et al., 2004; Zeebe and Archer, 2005). Therefore, and in view of its negative ecological  
41 and climatic side effects (Chisholm et al., 2004; Fuhrmann and Capone, 1991), artificial ocean fertilisation  
42 can practically be ruled out as a useful mitigation option. In any case, once it has re-entered the oceanic  
43 water column, the artificially stored carbon will reappear at the ocean surface – except for a small fraction  
44 within in each mixing cycle that becomes neutralized by CaCO<sub>3</sub> sediment dissolution – on a timescale of  
45 300-1000 years (Caldeira et al., 2005). Any deliberate ocean CO<sub>2</sub> storage, therefore, would remain a  
46 temporary anthropogenic feedback to rising atmospheric CO<sub>2</sub> concentrations.

#### 7.3.2.3 *Feedbacks at the glacial-interglacial scale*

49 From ice core measurements, it has been shown that the global atmospheric concentrations of the greenhouse  
50 gases CO<sub>2</sub> and CH<sub>4</sub> have changed with glacial-interglacial temperature and climate variations (see paleo  
51 climate carbon Box 6.23 and Sections 6.3.3. and 6.5.3 in Chapter 6; Monnin *et al.*, 2001). The changes were  
52 in a direction towards an amplification of the cooling from interglacial to glacial values starting at about 280  
53 ppm and 700 ppb decreasing to 190 ppm and 450 ppb respectively. The negative trend in CH<sub>4</sub> values with  
54 temperature is attributed mainly to changes in the terrestrial vegetation with a reduction of wetlands and an  
55 increase in permafrost areas. The lower glacial CO<sub>2</sub> values must be caused by marine processes as the  
56 reduction of terrestrial vegetation under colder and dryer conditions would have caused a relative increase of  
57 atmospheric CO<sub>2</sub> (Kaplan *et al.*, 2002). In order to compensate for this terrestrial carbon release, even a

1 somewhat larger drawdown of atmospheric CO<sub>2</sub> than 90 ppm must be explained by oceanic processes. These  
2 have not yet been conclusively identified. Therefore, a significant gap in our knowledge of ocean carbon  
3 cycle climate feedbacks still remains (Archer *et al.*, 2000; Sigman and Boyle, 2000). As in the glacial ocean,  
4 we expect a slowing down of oceanic overturning during a future climate change. The mode of ocean  
5 circulation, e.g. differences in shallow and deep circulation, may be considerably different from the glacial  
6 situation. Also the ca. 120 m interglacial-glacial sea level lowering may have led to a considerably different  
7 biogeochemical forcing of the open ocean than today. For these reasons, the paleo-record may not be an  
8 overall reliable analog for the future evolution of carbon cycle feedback processes.  
9

### 10 **7.3.3 Interannual and Regional Variation in Carbon Fluxes Inferred from Observations**

#### 11 *7.3.3.1 Global budget - interannual fluctuations*

12 The observed rate of atmospheric CO<sub>2</sub> increase exhibits large variations every 2-4 years (see Figure 2.4),  
13 which cannot be accounted for by the small year-to-year variability (IAV) in global fossil CO<sub>2</sub> emissions.  
14 This implies the existence of year-to-year fluctuations in the CO<sub>2</sub> uptake by land and ocean regions. Over the  
15 past two decades, low CO<sub>2</sub> uptake rates (i.e., high atmospheric growth rates) occurred in 1987, 1994–1995,  
16 1997–1998, and in 2002–2003. The record lowest uptake of 0.1 Pg-C only (highest atmospheric growth rate  
17 of 2.7 ppm) occurred in 1998 during the strongest El Niño of the 20th century. The yearly increase in  
18 atmospheric CO<sub>2</sub> in 2002 and 2003 exceeded for the first time 2 ppm yr<sup>-1</sup> in two consecutive years, while  
19 there was only a rather weak El Niño. Conversely, high CO<sub>2</sub> uptake (low growth rates) occurred in 1981,  
20 1992–1993 and 1996–1997. Generally, the high and low CO<sub>2</sub> uptake periods correspond to the succession of  
21 La Niña and El Niño episodes, as has long been recognized (Bacastow, 1976), although the relationship  
22 between CO<sub>2</sub> growth and the Southern Oscillation Index is evolving with time (Lintner, 2002). There are at  
23 least two remarkable exceptions to this, the 1992–1993 high uptake period popularly called the "post  
24 Pinatubo" period, and 2002–2003 low uptake period where for the first time since observations began, low  
25 uptake did not coincide with any developing El Niño.  
26

27  
28 [INSERT FIGURE 7.3.8 HERE]  
29

30 Since the TAR, a consensus has emerged from top-down inversion studies that the CO<sub>2</sub> fluxes IAV is mostly  
31 due to land fluxes, and that the Tropical lands contribute strongly to the global terrestrial IAV (Figure 7.3.8).  
32 Such a predominant terrestrial IAV forcing is indicated from: (1) CO<sub>2</sub> concentrations observed at different  
33 stations and used in atmospheric inversions by Bousquet *et al.* (2000), Rödenbeck *et al.* (2003) and Baker *et al.*  
34 (2004); (2) consistent relationships between δ<sup>13</sup>C and CO<sub>2</sub> globally (Francey *et al.*, 2001) or zonally, and  
35 (3) direct measurement of O<sub>2</sub> and CO<sub>2</sub> since 1991 (see Figure 2.2; Manning, 2001; Manning and Keeling,  
36 2005).  
37

#### 38 *7.3.3.2 Bottom-up estimates of ocean atmosphere carbon fluxes from observations*

39 Due to the high variability of air-sea CO<sub>2</sub> fluxes, the still scarce direct measurements of sea surface pCO<sub>2</sub> and  
40 the varying wind speeds, it is difficult to firmly assess changes in the carbon flux between ocean and  
41 atmosphere over the past decades (see Chapter 5, Section 5.4.2.2). Takahashi *et al.* (2002) normalize all  
42 available sea surface pCO<sub>2</sub> measurements between 1956 and 2000 for the reference year 1995 to come up  
43 with an annual oceanic net uptake of about 2.2 (±0.5) Pg C yr<sup>-1</sup>. This number may be biased due to special  
44 wind speed conditions in 1995 and errors in the height of the wind speed data applied. Sabine *et al.* (2004)  
45 estimate the total oceanic uptake of anthropogenic CO<sub>2</sub> to be 118(±19) Pg C for the period 1800–1994 from  
46 inorganic carbon measurements and a tracer-based technique for separation of anthropogenic carbon from  
47 natural carbon in the ocean water column. This indirect method can lead to an overestimate of the carbon  
48 uptake which can be corrected to 111(±25) Pg C for the period 1800–1994 (Keeling, 2005). Forward model  
49 simulations with a biogeochemical ocean general circulation model forced by daily NCEP/NCAR reanalysis  
50 data from 1948–2003 yield an oceanic net CO<sub>2</sub> uptake of 1.49 Pg C yr<sup>-1</sup> for the years 1980–1989 and 1.74 Pg  
51 C yr<sup>-1</sup> for 1990–1999 (Wetzel *et al.*, 2005). From the beginning of the industrial revolution up to now, the  
52 Southern Ocean turned from a source to a net sink of atmospheric CO<sub>2</sub> (Hoppema, 2004). The North Atlantic  
53 CO<sub>2</sub> sink strength is weakening in recent years (Lefèvre *et al.*, 2004). ENSO events and decadal variability  
54 have a strong impact on the net marine CO<sub>2</sub> uptake in the Pacific Ocean (Takahashi *et al.*, 2003; Wetzel *et al.*,  
55 2005).  
56

#### 1 7.3.3.4 *Regional carbon fluxes inferred from bottom-up approaches*

##### 2 7.3.3.4.1 *Terrestrial observations*

3 Locally, stand-level biometric measurements can be used to estimate the carbon balance of land ecosystems  
4 (Net Biome Productivity or NBP) but they are difficult to upscale since one must account for soil carbon  
5 changes and for the impact of disturbances in controlling the long term averaged carbon balance (Schulze et  
6 al., 2000; Korner, 2003). In boreal forests for example, carbon losses caused by disturbances and  
7 consumption of forest products comprises about 20% of NPP and can exceed the ecosystem carbon sink  
8 (Shvidenko and Nilsson, 2003). One cannot simply scale NBP from limited duration (less than 10 years)  
9 eddy-covariance NEE data (Baldocchi et al., 2001) because most eddy flux towers are located in young or  
10 middle-aged stands and do not sample the effect of disturbances, and because the length of those records is  
11 too short to average out the large fluctuations in NEE generally observed from one year to the next. The  
12 alternative to measure stand NBP is to use chronosequences (e.g., Mund et al., 2002).

13  
14 Regionally, extensive forest biomass inventory measurements estimate the measured volume growth of stem  
15 wood and changes in forest area, the modelled fine litter and coarse debris production, corrected for harvest  
16 and slash production in managed forests using bookkeeping type of models and other information for  
17 upscaling. The advantage here is covering and understanding spatial variability, at the expense of detailed  
18 knowledge of component processes. Systematic forest biomass inventories have been developed in temperate  
19 and boreal forests in North America, Western Europe, Russia, China to measure the commercial value of  
20 wood. In the tropics where there is no commercial forest inventory, stem growth measurements have been  
21 carried out as part of research networks (Malhi et al., 2004; Phillips et al., 1998).

##### 22 23 7.3.3.4.2 *Terrestrial models*

24 Many spatially explicit models of terrestrial ecosystems have been developed to analyze and predict the  
25 response of terrestrial carbon pools and fluxes to changing climate and atmospheric composition, and more  
26 recently to land use and management practices (e.g., McGuire et al., 2001). These models, similar to those  
27 used in the TAR, encapsulate biogeochemical processes responsible for biomass production as driven by  
28 climate and radiation, and calculate NPP as the difference between photosynthesis and autotrophic  
29 respiration components. After mortality, litter and soil organic matter decomposition is calculated, allowing  
30 estimates of the ecosystem carbon balance. The development of more realistic process-driven models  
31 enables the handling of diverse resolutions and scales, and the use of assimilation methods to allow the  
32 efficient processing of large amounts of data, particularly when using remote sensing observations (e.g.  
33 Wang and McGregor, 2003). The current generation of global terrestrial models are generally very crude in  
34 their description of 1) forest growth and management, 2) carbon cycling over cultivated lands and wetland,  
35 3) soil organic matter decomposition, 4) frozen soil carbon dynamics, 5) the effects of land-use change and  
36 fire, 6) coupling between carbon and nutrients cycling, 7) absorption of radiation by canopies, 8) effects of  
37 biodiversity.

##### 38 39 7.3.3.4.3 *Attribution of bottom-up regional carbon fluxes to processes*

40 It is fair to say that there are problems of quantifying NBP at various scales, and attributing it to the  
41 controlling processes. Firstly, at the spatial scale of stands, NBP can only be defined from the in situ carbon  
42 balance and thus does not include the fluxes of “displaced” carbon such as the losses to streams and rivers,  
43 or the fate of wood products in harvested forests. A second issue deals with temporal scales, because NBP  
44 contains “background” fluxes such as the formation of charcoal in fire-disturbed forests, the inclusion of  
45 carbon into mineral soil horizons, or the entrainment of rock-weathered DIC to the ocean. Those fluxes are  
46 part of the “background” natural carbon cycle and evolve on long time scales, whereas we are interested in  
47 NBP as the anomalous terrestrial uptake occurring on decadal time scales in response to the current  
48 perturbation of the carbon cycle. It is virtually impossible to separate “background natural” from  
49 “anthropogenic” NBP. In field studies, except maybe when using the bomb-radiocarbon signal to determine  
50 the soil component of ecosystem carbon storage (e.g., Trumbore et al., 1996). Ranges of NBP estimates for  
51 three distinct biomes are presented in Figure 7.3.9.

52  
53 [INSERT FIGURE 7.3.9 HERE]  
54

### 7.3.3.5 Regional carbon fluxes inferred from atmospheric CO<sub>2</sub> observations and inverse modelling

#### 7.3.3.5.1 Background

The atmosphere mixes and integrates surface fluxes that vary spatially and temporally. The distribution and temporal evolution of CO<sub>2</sub> in the atmosphere can be used to quantify surface fluxes, using numerical models of atmospheric transport combined with a representation of surface exchange fluxes. Typically the sub-model defining surface flux is adjusted to provide the best match to observed concentrations. This approach (“inverse modelling”) may be adapted to the specific circumstances at the scale of continents or large ocean gyres, where the precision and representativeness of the measurements should match that of the relevant major source and sink processes. Because of the long atmospheric life time of CO<sub>2</sub>, the horizontal gradients of concentrations which carry the information on the magnitude and spatial distribution of sources and sinks are quite small, and remain difficult to define with the present network of stations. In addition, atmospheric transport models are imperfect, to a largely unknown extent, and the inverted fluxes consequently depend on the model used.

#### 7.3.3.5.2 Atmospheric measurements

Atmospheric transport mixes air on the global scale on time scales of roughly a year. Therefore, the gradients in atmospheric CO<sub>2</sub> concentrations associated with regional fluxes are small compared to the background value of CO<sub>2</sub>. The global network of atmospheric CO<sub>2</sub> concentration measurement sites is shown in Chapter 2 (see also WMO/GAW, 2000). Most of these sites are discrete sampling sites, where flasks are collected approximately once a week, and there are a small number of in situ continuous stations. The network consists of different national networks and programs, with the US NOAA/CMDL contributing the largest dataset. The atmospheric network is much denser over the oceans than over the continents, and much denser in the Northern mid-to-high latitudes than in the Tropics, or in the Southern high latitudes. Fluxes upwind of areas with denser coverage of stations are better constrained by the data.

#### 7.3.3.5.3 Atmospheric inversion models

Using as an input the atmospheric CO<sub>2</sub> data described above, the atmospheric inverse models determine a global distribution of surface CO<sub>2</sub> sources and sinks which minimize a mismatch between modelled and observed gradients of concentrations, accounting for measurements errors. Bayesian synthesis inversions, the most widely used method pioneered by (Enting et al., 1995) are required simultaneously to fit optimally the available data while simultaneously preserving information present in an *a priori* flux model. When doing so, a number of modes of variability of the fluxes are explicitly solved for utilizing the information contained in the atmospheric observations, while a number of other modes are constrained by the *a priori* settings. One can explicitly propagate errors on atmospheric measurements and errors on the prior fluxes onto the sought fluxes, thus delivering a very useful set of uncertainty estimates. Generally, Gaussian random errors are assumed for the measurements and for the *a priori* fluxes, translating into Gaussian errors on the inverted fluxes. We report this *random uncertainty* as colored bars in the inversion fluxes of Figure 7.3.10.

[INSERT FIGURE 7.3.10 HERE]

In addition, inverse model results are biased by a certain choice of arbitrary settings. Those biases are difficult to quantify, and even harder to relate to the unknown truth. Arbitrary settings which can bias inverted fluxes include 1) the choice of a particular atmospheric transport model, 2) the assumed ability for this model to match a point observation with a grid-box model simulation (representation error), 3) the choice of fixed *a priori* temporal flux variations (e.g., as in Gurney et al., 2002; Gurney et al., 2003) or spatial flux variation within a given region (see discussion in Kaminski et al., 2001). There is also a bias arising from the fact that fossil fuel emissions are generally considered to be of perfectly known magnitude and patterns, so that their effect can be easily modelled and subtracted from atmospheric CO<sub>2</sub> data to solve for “residual” land and ocean fluxes. These various biases translate into a spread in the inverted fluxes. A lower limit to the magnitude of these biases can be obtained by performing an ensemble of inversions where the arbitrary settings are varied (e.g., using different transport models through the same inversion procedure, as done by the TRANSCOM-3 project ref). We report the *spread of mean fluxes* deduced from inversion ensembles with different settings as a grey bar in Figure 7.3.10.

The fluxes inferred from inverse models also depend critically on how the atmospheric observations are treated. Inhomogeneities and gaps in the data are inevitable, and the increasing number of sites in the network generates spurious changing estimates of regional fluxes as an artefact of adding stations downwind

1 of regions previously not well constrained. Further, network inhomogeneities, coverage denser over oceans  
2 (Patra et al., 2005), and calibration are also sources of bias on regional fluxes inferred from inversions.  
3 Several studies showed that the network is not dense enough for stations to be redundant. That means that in  
4 some cases, adding or removing sites (Bousquet et al., 1999; Law et al., 2003), or varying errors on the data  
5 (Krakauer et al., 2004) have strong implications for the inverted fluxes.

#### 7 7.3.3.5.4 Atmospheric inversion results: mean regional fluxes

8 In general, confidence in the long-term mean fluxes is lower than confidence in the year-to-year variations of  
9 the fluxes. Many inversion ensembles, each with different settings, have been produced since the TAR  
10 (Baker et al., 2004; Gurney et al., 2002; Gurney et al., 2003; Peylin et al., 2002, 2005a; Rödenbeck et al.,  
11 2003). An updated breakdown of oceans and land fluxes for large latitude bands is given in Figure 7.3.10.  
12 Inversion results are reported with their random estimation errors and a necessarily imperfect estimate of the  
13 (range of) bias implied by various settings in each ensemble. In the TRANSCOM-3 project 16 different  
14 transport models were compared through the same inverse procedure (Gurney et al., 2002, 2003). The results  
15 showed that differences between transport models cause uncertainties of smaller (or similar) magnitude than  
16 the random estimation error. This means that, for most regions, adding new measurements would be most  
17 beneficial for inversions. Over northern land regions, (e.g., boreal Asia), the models differ strongly in the  
18 way they respond to the seasonal cycle of the terrestrial CO<sub>2</sub> exchange, so that improving models is needed.  
19 Tropical regions, in particular tropical landmasses, could not be adequately constrained because of  
20 insufficient data, a general feature of all current inversions.

21  
22 All possible inversion settings were certainly not explored in the TRANSCOM-3 ensembles. In particular  
23 there was no assessment of the effect of the 'a priori aggregation' bias resulting from setting a fixed spatial  
24 pattern to the fluxes within each region. Inverting the fluxes at a much higher spatial resolution, up to the  
25 model grid itself (Kaminski et al., 1999; Rödenbeck et al., 2003). Peylin et al., 2005b) overcomes that  
26 limitation, but it implies nevertheless that the a priori flux model still influences the inversion result. Figure  
27 7.3.10 shows differences above their 1-σ errors between TRANSCOM-3 and Rödenbeck et al. (2003)  
28 ensembles over tropical and northern continents for the period 1992–1996. Part of these differences can be  
29 explained by different methodologies, in particular a different network of 35 stations (monthly flask data) in  
30 Rödenbeck et al., (2003) vs. 76 stations (smoothed flask data) in TRANSCOM-3. Figure 7.3.11 gives a  
31 range of 0.4–2.3 Pg-C y<sup>-1</sup> for uptake in Northern continents, and a range of 0.4 Pg-C y<sup>-1</sup> sink to 1.8 Pg-C y<sup>-1</sup>  
32 source over tropical continents during 1992–1996. Compensation effects obviously exist between the  
33 inversion results in the North and in the Tropics. Note that the inverse results for the 1992-1996 time period  
34 appear to confirm the anomalously strong uptake of CO<sub>2</sub> by terrestrial systems, although in some cases  
35 information on O<sub>2</sub> or δ<sup>13</sup>CO<sub>2</sub> was used in the inversion.

36  
37 [INSERT FIGURE 7.3.11 HERE]

38  
39 Figure 7.3.11 gives the breakdown of inversion fluxes for five large regions of the Northern Hemisphere:  
40 North America, North Atlantic, Europe, North Asia, and North Pacific. There are important differences in  
41 the mean of different ensembles, except over North America where all inversion fluxes generally agree  
42 within their errors to a mean sink ranging between 0.6 and 1.1 Pg-C y<sup>-1</sup>, although large uncertainties still  
43 pertain to that estimate (full range 0 to 1.6 Pg-C y<sup>-1</sup>). Larger differences between the inversion ensembles are  
44 found over Europe (range -0.9 to +0.2 Pg-C y<sup>-1</sup>) and over North Asia (range -1.2 to +0.3 Pg-C y<sup>-1</sup>), a latter  
45 region where inversions are sensitive to transport model choices, as seen from the large grey error in Figure  
46 7.3.11.

47  
48 *What is robust? i.e., inversion ensembles agree within their errors*

- 49 • The ocean uptake deduced from all inversions is higher in the Northern Extratropics than in the  
50 Southern Extratropics.
- 51 • The tropical oceans are outgassing CO<sub>2</sub> to the atmosphere (range 0 to 1.5 Pg-C y<sup>-1</sup>), as expected  
52 from upwelling of CO<sub>2</sub>-rich waters (Feely et al., 1999).
- 53 • The global land vs. ocean flux partitioning from inversions is within the uncertainty of the O<sub>2</sub>  
54 derived global budgets. This result cannot serve as an independent verification since some inversions  
55 precisely use that constraint.
- 56 • The Northern Hemisphere land fluxes are better constrained than the tropical land fluxes.

- There is a mean terrestrial sink in the Northern Hemisphere, with a fraction in North America, in Europe and Northern Asia. Very large uncertainties pertain to individual regional flux estimates, but the smallest spread is found over North America.
- The North American carbon sink estimated by recent inversions is large, but on average lower than an earlier, widely cited study (Fan et al., 1998). Nevertheless the estimate by Fan et al. (1998) remains within the inversion range of uncertainty.

*What is not robust? i.e., inversion ensembles disagree within their errors*

- The partition of terrestrial fluxes between northern and tropical lands.
- The sink function of tropical lands: if tropical lands inverted fluxes give a CO<sub>2</sub> source of similar magnitude than tropical deforestation in Table 7.3.2, then tropical undisturbed terrestrial ecosystems are about carbon neutral; if tropical lands inverted fluxes are a net sink of CO<sub>2</sub>, then undisturbed tropical ecosystems are a huge sink of CO<sub>2</sub>.
- The mean flux estimate of most individual regions is not robust through inversions.

#### 7.3.3.5.5 Mean regional fluxes, bottom-up estimates and inversions

Bottom up estimates of regional carbon fluxes have been summarized in Section 7.3.1.4.4. Comparing inversion estimates of large-scale sources and sinks with bottom up estimates is not an easy task because 1) inversion fluxes may already contain a certain amount of *a priori* knowledge of bottom-up fluxes, especially of air-sea fluxes, so that the two approaches are not fully independent 2) the time period for which inversion models and bottom-up estimates are available is often not consistent, in the presence of fluxes that can vary substantially from one year to the next (see 7.3.1.4.5), 3) the distribution of CO<sub>2</sub> fluxes given by inversions does not match the distribution of carbon storage generally given by bottom up, in the presence of lateral fluxes. Lateral fluxes act to transport carbon away from where CO<sub>2</sub> is exchanged to or from the atmosphere, in such a manner that the regional changes in *carbon storage* have a geographically distinct distribution than the CO<sub>2</sub> fluxes (Tans et al., 1995; Sarmiento and Sundquist, 1993). Bottom up methods are measuring carbon storage changes, and inversions CO<sub>2</sub> fluxes. Lateral fluxes include the emissions of reduced carbon compounds (VOCs, CO, CH<sub>4</sub>) by ecosystems and anthropogenic combustion processes, transported and oxidized by OH chemistry (Enting and Mansbridge, 1991; Folberth et al., 2005; Suntharalingam et al., 2005), trade of wood and food products harvested from ecosystems (Ciais et al., 2005; Imhoff et al., 2004), and riverine transport of dissolved inorganic and organic carbon to the ocean (Aumont et al., 2001; Meybeck, 1987). The main results from comparison of bottom-up with inversion mean fluxes are:

- Inversions determine a higher ocean sink in the Northern extratropics than in the Southern extratropics, in contrast with oceanographic estimates (Takahashi et al., 2002; Takahashi et al., 1999), although new measurements of  $\Delta p\text{CO}_2$  in winter over the Southern Ocean may reconcile both estimates (Metzl et al., 1999).
- Inversions tend to estimate higher terrestrial carbon uptake over Northern Hemisphere lands than bottom up estimates (Nabuurs et al., 1997; Goodale et al., 2002; Pacala et al., 2001; Janssens et al., 2003; Shvidenko and Nilsson, 2003) (see Figure 7.3.11), but part of this discrepancy can be explained by laterally-transported carbon via reduced carbon compounds, rivers and food and wood trade.
- Inversions determine that tropical land regions are either close to neutral, or a sink of CO<sub>2</sub>, qualitatively in agreement with limited forest biomass changes measurements in the Amazon (Phillips et al., 1998; Malhi and Grace, 2000).

#### 7.3.3.5.6 Atmospheric inversion results: interannual changes in regional fluxes

Three inversion ensembles calculated the changes in monthly regional fluxes over the last two decades (Bousquet et al., 2000; Rödenbeck et al., 2003). As for mean regional fluxes, these studies report a random error and the error range of sensitivity inversions with different settings (set of atmospheric stations, atmospheric measurement errors, transport model, the choice, a priori flux constraints, spatial or temporal scales at which the fluxes are solved for).

Bousquet et al., (2000) used large regions and varied different inversion settings for the period 1980–1998. They found that the year-to-year regional flux changes could be more robustly inverted than their mean values.

Rödenbeck et al. (2003) used one transport model and an inversion at the resolution of model grid for the period 1982–2002, and changed different inversion settings. In particular, different stations from the CMDL network (see Figure 2.3) were used, from 11 sites up 35 sites, showing an impact on the inversion mean fluxes but less on broad regions variability. The effect of using interannually varying winds was concluded to have a relatively small impact on the fluxes variations.

Baker et al. () used large regions but 13 different transport models for the period 1988–2002. Their results suggest that the spread of transport models has a lesser impact on the year-to-year regional flux anomalies than on the mean fluxes (see 7.3.1.3.3). In other words, the changes of regional fluxes owe more to errors in atmospheric data, a priori constraints, than to inter-model differences.

*What is robust? i.e., inversion ensembles agree within their errors*

- At the scale of the globe, the IAV of land fluxes is larger than the one of ocean fluxes.
- At the scale of large latitude bands the IAV of land fluxes is larger than the one of ocean fluxes
- Tropical land fluxes exhibit on average a larger variability than temperate and boreal land fluxes, and their variability suggests unambiguously an El Niño forcing, with anomalous sources during El Niño episodes in 1982–1983, 1986–1987 and 1997–1998, and anomalous sinks during La Niña episodes.
- The 1997–1998 high CO<sub>2</sub> growth rate episode is attributed to an abnormal source in the Tropics, with a significant fraction of it in South East Asia, in good agreement with bottom up estimates (fires in Indonesian moist forests)
- The Tropical Pacific ocean is a region where the significance of the inversion results for year-to-year changes is high, in good agreement with bottom up estimates ( $\Delta p\text{CO}_2$  observations)

*What is not robust? i.e., inversion ensembles disagree within their errors*

- The 1992–1993 low CO<sub>2</sub> growth rate episode during the post Pinatubo climate disturbance is attributed either to the Northern mid latitudes by Bousquet et al. and to the Tropical lands by the two other ensembles
- At the scale of continents or ocean basins in the Northern Hemisphere, the errors increase and the significance of the inverted flux inter-annual changes is quickly lost.

*7.3.3.5.7 Year to year changes in regional fluxes, inversion and bottom-up*

- Globally, inversion results agree with independent ocean carbon model calculations on the rather small variability of air sea fluxes ( $\pm 0.5 \text{ Pg-C y}^{-1}$  between extremes), and conversely on to large variability of land fluxes ( $\pm 4 \text{ Pg-C y}^{-1}$  between extremes). Yet, inversions and ocean models differ on the geographic contributions to the year-to-year variability.
- In the Northern Extratropics (Figure 7.3.11), Baker et al. (year?) and Bousquet et al. (year?) obtain a significant variability in air sea fluxes, in contrast to Rödenbeck et al. (2003). On the one hand, over the North Atlantic ocean, Gruber et al. extrapolated a large variability comparable to Bousquet et al. (year?) (extremes of  $\pm 0.3 \text{ Pg-C y}^{-1}$ ) using data from the Bermuda station during 1984–2000. On the other hand, for that region McKinley et al. (2004) modelled a small variability (extremes of  $\pm 0.1 \text{ Pg-C y}^{-1}$ ) comparable to the one of Rödenbeck et al. (2003), and suggested that a larger variability could be an effect of the large region settings (ref). Note that all inversions are not strictly independent from bottom-up studies, as they include a priori information to constrain the air-sea flux variations, which may in turn influence their results.
- El Niño and La Niña episodes force respectively a large release and uptake of CO<sub>2</sub> over the tropical continents, in particular over the Amazon. Flux anomalies are large ( $2-3 \text{ Pg-C y}^{-1}$ ) and in general good agreement with many bottom-up biospheric model calculations. In the Equatorial Pacific ocean, all inversions obtain a flux variability which compares well in magnitude and timing with ocean models results (Le Quere et al., 2000, 2003; McKinley et al., 2003, 2004) and with  $\Delta p\text{CO}_2$  observations (Feely et al., 2002). The flux variability in that region is paced by El Niño, with a smaller net CO<sub>2</sub> source to the when upwelling of CO<sub>2</sub>-rich waters diminishes.
- The 1992–1993 low CO<sub>2</sub> growth rate episode during the post Pinatubo climate disturbance is not a robust result of inversions. Bousquet et al. (2000) find an enhanced terrestrial uptake in the Northern Hemisphere, while Baker et al. (year?) and Rödenbeck et al. (year?) place it in the Tropics. Analysis

of remotely-sensed vegetation activity and a terrestrial carbon model calculation by Lucht et al. (2002) produces good agreement with Bousquet et al. Apart from higher NPP, lower respiration or a combination of both conventionally expected from the cooling, an overall increase in photosynthesis may have occurred via the increased fraction of diffuse sunlight (Roderick and Farquhar, 2001). However, this increase is not supported by the changes in the NDVI or the seasonal cycle of CO<sub>2</sub> post-Pinatubo (Angert et al., 2004).

- The 1997–1998 high CO<sub>2</sub> growth rate period can be consistently explained by abnormally high and intense fire emissions. For Indonesia alone, a source of +0.8 to +2.6 Pg-C from peat forest fires was estimated by Page et al. (2002). For the globe, approximately 2/3 of the 1997–1998 growth rate excess was attributed to fire emission anomalies, with main contributors South East Asia (60%), South America (30%) and a small Siberian contribution (van der Werf et al., 2004). Similarly, Langenfelds et al. (2002) analyzed the correlations in the interannual growth rate of CO<sub>2</sub> and other species at 10 stations and linked the 1997–1998 (and the 1994–1995) anomalies and to high fires emissions as a single process. The relationship between El Niño and CO<sub>2</sub> emissions from fires is however not uniform, with for instance in Africa drier conditions determining less fire emissions due to less biomass. In addition, co-varying processes such as reduced NPP caused by the dry conditions over tropical forests during El Niño episodes may be superimposed on fire emissions.

### 7.3.4 Coupled Climate-Carbon Cycle Projections

#### 7.3.4.1 Introduction

The majority of model simulations of future climate prescribe future scenarios for global CO<sub>2</sub> concentrations using relatively simple offline carbon cycle models. Hence model projections do not account for two-way coupling between climate change and the carbon cycle. But we know that reservoirs of carbon on land and in the ocean respond to changes in climate. Global concentrations of atmospheric CO<sub>2</sub> have been observed to respond to the El Niño Southern Oscillation and Arctic Oscillation (Jones et al., 2001; Bousquet et al., 2000; Rayner et al., 1999; Lintner, 2002; Russell and Wallace, 2004) and to the perturbation of climate and aerosol scatter from the Pinatubo volcanic eruption (Hansen et al., 1996; Jones and Cox, 2001; Lucht et al., 2002; Angert et al., 2004).

The TAR reported two initial climate projections using AOGCMs with interactive carbon cycles. Both indicated positive feedback due largely to impacts of climate warming on land carbon storage (Cox et al., 2000; Friedlingstein et al., 2001), but the magnitude of the feedback varied markedly between the models (Friedlingstein et al., 2003). Since the TAR a number of other climate modelling groups have completed climate-carbon cycle projections (Thompson et al., 2004; Matthews et al., 2004a, b; Zeng et al., 2004; Brovkin et al., 2004; Raddatz et al., 2005; Fung et al., 2005; Kawamiya et al., 2005; Sitch et al., 2005), as part of the Coupled Climate-Carbon Cycle Model Intercomparison Project (C<sup>4</sup>MIP). The models involved in C<sup>4</sup>MIP differ in the complexity of their components (Table 7.3.4).

**Table 7.3.4.** Models involved in the coupled climate-carbon cycle intercomparison project (C<sup>4</sup>MIP).

Model	Atmosphere	Ocean	Land Carbon	DGVM	Ocean Carbon	Reference
HadCM3LC	HADCM3 2.5° × 3.75°, L19	2.5° × 3.75°, L20 flux-adjusted	MOSES/ TRIFFID	Yes	HadOCC	Cox et al., 2000
IPSL-CM2C	LMD5 5.6° × 3.6°, L19	OPA,	SLAVE	No	NPZD	Dufresne et al., 2002
NCAR CSM-1	CCM3 T31, L18	NCOM 3.6° long, 0.8–1.8° lat	LSM, CASA	No	OCMIP-biotic	Doney et al., 2005; Fung et al., 2005
MPI	ECHAM5, T63, L19	MPI-OM, 1.5°, L40,	JSBACH	Yes	HAMOCC5	Raddatz et al., 2005
LLNL	CCM3, 2.8° × 2.8°, L18	POP 0.6° × 0.6°, L40	IBIS	Yes	OCMIP	Thompson et al., 2004
FRCGC	CCSR/NIES/FRCGC T42I.20 (~2.8° ×	COCO No flux adjustment,	Sim-CYCLE	No	NPZD	Kawamiya et al., 2005:

	2.8°)	3-hour coupling, (0.5–1.4°) × 1.4°, L20				Hasumi & Emori, 2004
UMD	QTCM	One-box	VEGAS	Yes	Abiotic tracer	Zeng et al., 2004
UVic-2.7	EMBM 1.8° × 3.6°	MOM 2.2, 1.8° × 3.6°, L19, no flux adjustment	MOSES/ TRIFFID	Yes	OCMIP Abiotic	Meissner et al., 2003; Matthews et al., 2005
CLIMBER2- LPJ	2.5-D statistical- dynamical, 10° × 5.1°	Zonally-averaged; 2.5°lat, 3 basins	LPJ	Yes	NPZD	Brovkin et al., 2004; Sitch et al., 2005
BERN-CC	EBM 2.5° × 3.75°	HILDA box-diffusion model	LPJ	Yes	Perturbation approach	Joos et al., 2001; Gerber et al., 2003

The models were forced by historical and SRES A2 anthropogenic emissions of CO<sub>2</sub> for the 1850–2100 time period. Each modelling group carried out at least two simulations, one “coupled” in which climate change affects the carbon cycle, and one “uncoupled” in which CO<sub>2</sub> is treated as a non-radiatively active gas (so that the carbon cycle experiences no CO<sub>2</sub>-induced climate change). The difference between these runs defines the climate-carbon cycle feedback.

#### 7.3.4.2 Carbon budget changes

Table 7.3.5 shows the change in the carbon budgets over the course of the uncoupled and coupled runs, including of the fractions of the total CO<sub>2</sub> emissions which end up in the atmosphere, land and ocean in 2100.

**Table 7.3.5.** Simulated air, land, and ocean -borne fractions in 2100 of the cumulative anthropogenic CO<sub>2</sub> emissions over the entire period of the model runs. Columns 2–4 show reservoir inventory fractions as: coupled (uncoupled).

Model	Air-borne Fraction	Land-borne Fraction	Ocean-borne Fraction	Feedback Factor ( <i>F</i> )	Atmospheric CO <sub>2</sub> : Coupled – Uncoupled (ppm)
HadCM3LC	0.72 (0.49)	0.05 (0.30)	0.24 (0.20)	1.47	225
IPSL-CM2C	0.47 (0.40)	0.22 (0.30)	0.32 (0.30)	1.17	75
NCAR-CSM1	0.54 (0.52)	0.25 (0.26)	0.21 (0.22)	1.04	20
MPI	0.54 (0.46)	0.22 (0.30)	0.24 (0.24)	1.17	82
LLNL	0.41 (0.36)	0.44 (0.49)	0.15 (0.15)	1.14	54
FRCGC	0.63 (0.60)	0.10 (0.10)	0.27 (0.30)	1.05	25
UMD	0.64 (0.55)	0.01 (0.06)	0.35 (0.39)	1.16	87
UVic-2.7	0.59 (0.48)	0.17 (0.28)	0.23 (0.26)	1.23	127
CLIMBER	0.58 (0.52)	0.22 (0.27)	0.20 (0.21)	1.12	53
BERN-CC	0.48 (0.42)	0.26 (0.32)	0.26 (0.26)	1.14	75

The uncoupled models all take-up fairly similar fraction of emissions (airborne fraction 56±7 (1σ) %), but this is achieved through different contributions from the land and the ocean. The fraction of emissions taken-up by the land is 27±12% owing primarily to differences in how CO<sub>2</sub>-effects on photosynthesis are modelled. The change in oceanic inventory is 25±7% of the total emission, mostly due to differences in the transport of CO<sub>2</sub> to depth. In the models, there are compensating variations between land and ocean uptake, so the range in the atmosphere is relatively smaller than those in the land and ocean reservoirs.

The effect of climate change on the rate of increase of atmospheric CO<sub>2</sub> can be quantified by the feedback factor:  $F = \Delta C_A^c / \Delta C_A^u$ , where  $\Delta C_A^c$  is the change in CO<sub>2</sub> in the coupled run, and  $\Delta C_A^u$  is the change in CO<sub>2</sub> in the uncoupled run. All models show positive climate-carbon cycle feedbacks (Table 7.3.5), with  $F \approx 1.17 \pm 0.12$  but with widely varying magnitudes.

#### 7.3.4.3 Feedback analysis

In order to understand model behaviour and identify key uncertainties, model experiments are compared in terms of the responses of the land and ocean carbon uptake to changing climate and CO<sub>2</sub> (Friedlingstein et al., 2003):

$$\Delta C_L = \beta_L \Delta C_A + \gamma_L \Delta T$$

$$\Delta C_O = \beta_O \Delta C_A + \gamma_O \Delta T$$

where  $\Delta C_L$  and  $\Delta C_O$  are the change in land and ocean carbon storage (both in Pg-C) arising from an increase in atmospheric CO<sub>2</sub> concentration of  $\Delta C_A$  (ppmv) and a temperature increase of  $\Delta T$  (°C). The gain of the climate-carbon cycle feedback loop,  $g$ , can be defined in terms of the feedback factor,  $F$ :

$$F = 1 / (1 - g) = \Delta C_A^c / \Delta C_A^u$$

The effect of changing CO<sub>2</sub> on global mean temperature can be approximated as  $\Delta T \approx \alpha \Delta C_A$  where  $\alpha$  is the linearized transient climate sensitivity factor for rising CO<sub>2</sub> (units: °C ppm<sup>-1</sup>), leading to an expression for gain in terms of the sensitivity coefficients (Friedlingstein et al., 2003):

$$g = -\alpha (\gamma_L + \gamma_O) / (1 + \beta_L + \beta_O)$$

Note that the gain of the carbon cycle is larger for higher effective climate sensitivities  $\alpha$  (Jones et al., 2003a; Govindasamy et al., 2005) and for more negative values of  $\gamma_L$  and  $\gamma_O$ . However, the climate-carbon cycle feedback is weaker if ocean and land uptake respond very positively to increasing CO<sub>2</sub> (i.e., large  $\beta_L$  and  $\beta_O$ ).

Although this analysis neglects some of the non-linear aspects of the climate-carbon cycle feedback, it provides a valuable way to begin characterising the different model responses. Table 7.36 compares the  $\alpha$ ,  $\beta$  and  $\gamma$  coefficients from each of the C<sup>4</sup>MIP models.

**Table 7.3.6.** Carbon cycle gain ( $g$ ) along with component sensitivities of climate to CO<sub>2</sub> ( $\alpha$ ), and land and ocean carbon storage to CO<sub>2</sub> ( $\beta_L$ ,  $\beta_O$ ) and climate ( $\gamma_L$ ,  $\gamma_O$ ).

Model	$\alpha$ (°C ppm <sup>-1</sup> )	$\beta_L$ (Pg-C ppm <sup>-1</sup> )	$\beta_O$ (Pg-C ppm <sup>-1</sup> )	$\gamma_L$ (Pg-C °C <sup>-1</sup> )	$\gamma_O$ (Pg-C °C <sup>-1</sup> )	Gain $g$
HadCM3LC	0.0054	1.3	0.9	-175	-24	0.31
IPSL-CM2C	0.0064	1.6	1.6	-98	-30	0.15
NCAR-CSM1	0.0033	1.1	0.9	-22	-16	0.04
MPI	0.0069	1.4	1.1	-63	-22	0.20
LLNL	0.0077	2.8	0.9	-85	-14	0.10
FRCGC	0.0063	0.3	1.1	0	-23	0.04
UMD	0.0050	0.2	1.5	-40	-67	0.14
UVic-2.7	0.0059	1.2	1.1	-100	-42	0.20
CLIMBER	0.0053	1.1	0.9	-57	-22	0.10
BERN-CC	0.0046	1.6	1.3	-104	-38	0.13

(1) *Increase in ocean carbon uptake with atmospheric CO<sub>2</sub>.* The ocean will take up CO<sub>2</sub> at a rate which depends on the difference between the partial pressures of CO<sub>2</sub> in the atmosphere and the surface ocean.

1 Model estimates of uptake differ primarily because of differences in the rate at which carbon is exported  
2 from the surface ocean to depth by both the large-scale circulation (Doney et al., 2004) and the biological  
3 pump (Sarmiento et al. 2004). The coefficient  $\beta_0 \approx 1.14 \pm 0.26$  Pg-C ppmv<sup>-1</sup> gives the rate of change of ocean  
4 carbon storage with CO<sub>2</sub> alone.

5  
6 (2) *Increase in land carbon uptake with atmospheric CO<sub>2</sub>*. Although the experimental evidence of CO<sub>2</sub>  
7 fertilisation at the patch-scale is equivocal (Oren et al., 2001; Luo et al., 2004) most climate-carbon cycle  
8 models represent the current land-carbon sink by this mechanism (Cramer et al., 2001). Figure 7.3.12a shows  
9 the change in land carbon storage against atmospheric CO<sub>2</sub> concentration from each of the uncoupled C<sup>4</sup>MIP  
10 runs. The mean gradient of the line defines  $\beta_L \approx 1.24 \pm 0.73$  Pg-C ppmv<sup>-1</sup>. Table 7.3.6 shows the range of  $\beta_L$ .  
11 Experiments with the LLNL model, in which the CO<sub>2</sub>-fertilisation effect was capped at current day show the  
12 extent to which a large  $\beta_L$  can reduce the atmospheric CO<sub>2</sub> levels and consequently the magnitude of the  
13 carbon cycle feedback (Thompson et al. 2004).

14  
15 [INSERT FIGURE 7.3.12 HERE]

16  
17 (3) *Dependence of ocean carbon uptake on climate*. Climate change can affect ocean carbon uptake by  
18 influencing the transport of carbon to depth by the large-scale circulation (e.g., by slowing down the  
19 thermohaline circulation), the solubility pump and the biological pump. In particular, increases in thermal  
20 stratification as the upper ocean warms may suppress vertical transport. The  $\gamma$  factor for the ocean is defined  
21 as:  $\gamma_O = (\Delta C_O - \beta_O \Delta C_A) / \Delta T$ , where  $\beta_O$  is defined from the uncoupled run as discussed above. Table 7.3.6  
22 shows the values of  $\gamma_O \approx -31 \pm 15$  Pg-C °C<sup>-1</sup>, with all models showing a suppression of ocean uptake with  
23 climate change.

24  
25 (4) *Dependence of land carbon storage on climate*. Land carbon storage depends on the balance between the  
26 input of carbon as Net Primary Productivity (NPP), and the loss of carbon as heterotrophic (soil) respiration  
27 ( $R_h$ ). Both of these terms are strongly climate dependent. Plant productivity depends on, inter alia, water  
28 availability and ambient temperatures. Changes in water availability depend critically upon uncertain  
29 regional aspects of climate change projections and are therefore likely to be a dominant source of uncertainty  
30 (see Chapter 11).

31  
32 The overall sensitivity of land carbon storage to climate (Figure 7.3.12b) is quantified in terms of  
33  $\gamma_L \approx -73 \pm 50$  Pg-C °C<sup>-1</sup>. Seven of the ten models have  $\gamma_L$  in the range -40 to -104 Pg-C °C<sup>-1</sup>, even though  $\gamma_L$   
34 ranges from 0 (FRCGC) to -175 Pg-C °C<sup>-1</sup> (Hadley). The models all simulate a reduction in land carbon  
35 storage due to climate change alone, primarily due to a reduction in photosynthesis as water stress increases,  
36 and an accelerating breakdown of soil organic matter as the land warms.

37  
38 Representation of photosynthesis in the models varies greatly in complexity, with some models including  
39 climate and biogeochemical control on stomatal conductance and hence canopy photosynthesis,  
40 transpiration, and leaf area; while others include highly simplified treatment of the dependence of canopy  
41 photosynthesis on climate. The wide range in photosynthesis response is also tied to the very different  
42 representation of hydrologic cycle in the models (physical climate model or carbon model). The well-  
43 documented range in modelled temperature and precipitation changes (see Chapter 10) thus present very  
44 different degrees of water stress for photosynthesis.

45  
46 The C<sup>4</sup>MIP models utilise different representations of soil carbon turnover, ranging from single-pool models  
47 (Hadley) to nine-pool models (NCAR- CSM1). However, all of the models assume a similar acceleration of  
48 decay with temperature, approximately equivalent to a doubling of the specific respiration rate for every 10  
49 °C warming (i.e.  $q_{10} = 2$ ). There is also a wide range of soil moisture sensitivity which, when coupled with  
50 the range of modelled soil moisture changes, contributes to the divergent results.

51  
52 The temperature sensitivity is broadly consistent with a long history of lab and field measurements (Raich  
53 and Schlesinger, 1992), although there is an ongoing debate about the extent of acclimation of  $R_h$  to higher  
54 temperatures. The expected dependence on temperature was not found at the whole-ecosystem level for  
55 decadal time scales, in forest soils (Giardina and Ryan, 2000, Melillo et al., 2002) grasslands (Luo et al.,  
56 2001), or boreal forests (Dunn et al., 2005). However, these discrepancies in part reflect the rapid  
57 acclimatization of labile pools of organic matter through changes in the pool size. Strong temperature

1 responses can still occur on much longer time scales so long as litter inputs to the soil are maintained (Knorr  
2 et al., 2005), *although this is a theoretical result yet to be demonstrated in the field.*

3  
4 The strong negative impact of climate change on land carbon storage in the Hadley model is the primary  
5 reason for its large positive climate-carbon cycle feedback (Friedlingstein et al., 2003). A number of studies  
6 have looked at aspects of this, including the marked drying under climate change in the Amazon basin (Cox  
7 et al., 2004), the choice of  $q_{10} = 2$  for soil respiration (Jones et al., 2003b), the use of a single-pool soil  
8 carbon model (Jones et al., 2005b), and the parameterisation of plant respiration (Huntingford et al., 2004).  
9 In each case the characteristics of the Hadley Centre model appear to encourage a larger  $\gamma_L$ , but no single  
10 assumption accounts completely for the large sensitivity, or has been unequivocally falsified. For example,  
11 the use of a single soil carbon pool has been suggested as the predominant reason for the large feedback  
12 strength in the Hadley model (Zeng et al., 2004), but recent tests with a multi-pool soil model indicate that  
13 this explains only a small fraction of the discrepancy (Jones et al., 2005b).

14  
15 The relatively small positive feedback in the NCAR CSM-1 model (small magnitude of  $\gamma_L$ ) reflects a trade-  
16 off between CO<sub>2</sub> sources from degraded tropical ecosystems and CO<sub>2</sub> uptake in higher latitudes (Fung et al.  
17 2005). The model indicates negative covariance between soil moisture and temperature (warmer, drier) in  
18 the tropics but positive correlation (warmer, wetter) at high latitudes (Doney et al., 2005 fig. 2; or Fung et al.  
19 (2005), Figure 7.3.13). Since evapotranspiration and vegetation demand for water increase in a warmer  
20 climate, tropical ecosystems degrade and high latitude systems agrade. All the models tend to show  
21 increasing aridity at low latitudes but give divergent results at high latitudes (e.g. Cox, Betts, Bertelot,  
22 Matthews, Fung), indicating that the response of the global system may need to be disaggregated in order to  
23 elucidate the driving factors in each model. The large magnitude for  $\gamma_L$  in the Hadley model reflects negative  
24 covariance in both low and high latitudes, as a result of the large climate sensitivity ( $\alpha$ ).

25  
26 [INSERT FIGURE 7.3.13 HERE]

27  
28 *Summary: Importance of carbon cycle-climate feedback* - The coupled climate-carbon cycle models  
29 participating in the C<sup>4</sup>MIP project all support the following qualitative statements:

- 30 • Climate change will increase the fraction of anthropogenic CO<sub>2</sub> emissions which remain airborne  
31 (see Table 7.3.5), producing a positive feedback on climate change.
- 32 • CO<sub>2</sub> increase alone will tend to enhance uptake by both the land and the ocean ( $\beta_O > 0$ ,  $\beta_L > 0$ ).
- 33 • Climate change alone will tend to suppress land and ocean carbon uptake ( $\gamma_O < 0$ ,  $\gamma_L < 0$ ).

34 However, there is much less agreement over the magnitude of these various effects. Seven out of the ten  
35 models produce a feedback factor in the range 1.11 to 1.24, but they do this for very different reasons.  
36 Furthermore, it is not currently possible to rule out the large feedback ( $F=1.44$ ) in the Hadley Centre model,  
37 or the very small feedback in the NCAR-CSM1 model ( $F=1.03$ ). At this stage it is safest to assume that an  
38 amplification of CO<sub>2</sub> increase by 2100 in the range 10–25% is most likely, but with larger positive feedbacks  
39 possible (as a high-impact, low probability case). All models point towards declines of tropical ecosystems, a  
40 potentially major impact on world biotic and economic resources. But model validation is at an early stage,  
41 and issues of the first order remain unresolved. Thus the potentially important results from this very first  
42 C<sup>4</sup>MIP should be regarded as preliminary indicators of matters needing intensive study.

#### 43 44 7.3.4.4 *Missing processes and critical uncertainties*

45 The model intercomparison was designed as a benchmark study, rather than a simulation or prediction.  
46 While the results for the 19<sup>th</sup> and 20<sup>th</sup> centuries resemble those observed, they cannot be directly compared  
47 with the real world, as critical mechanisms (e.g. solar variability, other anthropogenic radiative forcing,  
48 legacies of prior land use, macro- and micro-nutrient limitation on terrestrial and marine productivity) were  
49 excluded from the experimental protocol. The results from this intercomparison highlight a number of  
50 critical outstanding issues for understanding climate-carbon cycle feedback.

51  
52 Differences in model estimates of oceanic uptake also arise because of the differences in land uptake, with  
53 the oceans steadily absorbing carbon not absorbed by the land. Models with the most efficient export to the  
54 deep tend to have less negative  $\gamma_O$  and larger  $\beta_O$ —viz. oceanic sequestration is relatively efficient and is only  
55 slightly slowed down by climate warming;  $g$  is consequently reduced. These factors are discussed in 7.3.2.2.

56  
57 On land, major uncertainties in the projections are the following:

1  
2 (1) *Dependence of carbon storage on temperature and soil moisture.* The global inventory of fossil fuel  
3 carbon that ends up on land is the result of local competition between temperature and moisture effects on  
4 photosynthesis and respiration, and the regional competition between regions of carbon accumulation and  
5 loss. Spring warming would lengthen growing season, especially at middle to high latitudes (e.g., Keeling et  
6 al., 1996; Myneni et al., 1997), while summer drying or tropical droughts would decrease net carbon uptake  
7 (Angert et al., 2005). Thus the wide range and hence uncertainties in projections of land carbon storage are  
8 intimately tied not only to the sensitivity of these processes to climate change, but also to uncertainties in the  
9 modelled projections of climate change itself. Separating moisture dependence from temperature  
10 dependence of photosynthesis and decomposition is critical but not straightforward, because the co-variance  
11 of temperature and moisture anomalies varies spatially and temporally. Furthermore, as recognized early by  
12 Jenny (1941), rates of mineralization of the large pools of soil organic matter depend at least as much on soil  
13 moisture as on temperature. However, experimental evidence is equivocal at best at the whole-ecosystem  
14 level for decadal time scales. These anomalies suggest that climate models may underestimate positive  
15 feedback of climate on photosynthesis, and overestimate positive feedback of climate on mineralization of  
16 soil carbon for the year 2100.

17  
18 (2) *CO<sub>2</sub> fertilization and land use legacy.* The C<sup>4</sup>MIP climate-carbon cycle models represent the current  
19 land-carbon sink as stimulated carbon assimilation and sequestration in response to rising CO<sub>2</sub>. Experimental  
20 evidence for this “CO<sub>2</sub> fertilization” is also equivocal at best (Cramer et al., 2001; Oren et al., 2001; Luo et  
21 al., 2004; DeLucia et al., 2005). The models exclude, by design, the effects of legacies of prior land use, in  
22 contrast to current hypothesis/observation that the contemporary forest landscape, with mostly young ages,  
23 plays a major role in current net carbon sequestration (e.g., Pacala et al., 2001; Schimel et al., 2001; Hurtt et  
24 al., 2002). Thus the present models might overestimate  $\beta_L$  and underestimate positive feedback by  
25 confounding legacies of land use, which do not increase with rising CO<sub>2</sub>, with fertilization, which does  
26 respond to CO<sub>2</sub>.

27  
28 (3) *Soil carbon turnover.* The C<sup>4</sup>MIP models specify, for each of the different soil carbon pools, “standard”  
29 turnover rates for a specified temperature and soil moisture, with the rates modulated by variations in  
30 temperature and moisture. Recent <sup>14</sup>C measurements in temperate ecosystems (check) show that over half of  
31 the photosynthetic flux is returned to the atmosphere within 5 years via heterotrophic respiration, so that the  
32 flux-weighted turnover time of soil carbon is shorter than the mass-weighted turnover time (Trumbore and  
33 Gaudinski, 2003). A shorter turnover time would decrease carbon storage and increase the positive feedback  
34 on climate. On the other hand, manipulation experiments have also shown changes in the microbial  
35 population at high CO<sub>2</sub> levels, so as to slow soil carbon turnover (Treseder et al., 2003), increase carbon  
36 storage and reduce positive feedback on climate.

37  
38 (4) *Limits to growth.* Nitrogen and other resources are known to limit photosynthesis. Other limitations  
39 include structural limitations and genetic longevity (Zavaleta et al., 2003a, b), which are not yet included in  
40 this generation of models. These limits would mostly likely decrease the unrestrained carbon uptake and  
41 storage in the C<sup>4</sup>MIP models and increase  $|\gamma_L|$ , thus magnifying the carbon-climate feedback.

42  
43 (5) *Other ecosystem processes.* Conversion of biomass and soil organic matter to CO<sub>2</sub> also depends on  
44 combustibility of ecosystems and organic soils (Kasischke et al., 2005; Cochrane, 2003; Nepstad et al., 2004;  
45 Randerson et al., 2002a-d, 2005; Jones and Cox, 2005), not just on respiration. The correlation between  
46 temperature and soil moisture shown in Figure 7.3.13 suggests disturbance regimes not included in the  
47 C<sup>4</sup>MIP models would likely further magnify carbon-climate feedbacks.

48  
49 (6) *Evolution of the landscape.* Changes in the distribution of ecosystems would occur with changes in  
50 climate and atmospheric CO<sub>2</sub> levels, at the same time that humans are expected to alter the landscape. Yet  
51 the 21<sup>st</sup> century climate and atmospheric conditions have no analogue in past climates. It is difficult to assess  
52 the full suite of carbon-climate consequences of these changes, other than to assert that conversion from high  
53 to low biomass would tend to increase the positive carbon-climate feedback, and vice versa.

54  
55 In order to reduce the uncertainties in climate-carbon cycle projections, it is critically important to constrain  
56 carbon cycle models by observations. This requires long-term measurements of natural ecosystems,  
57 ecosystem manipulations covering climatically relevant time scales, and more sophisticated models which

1 can more easily utilise this data. The current simplistic representations of ecosystem processes run the risk of  
2 misidentifying the causes of contemporary carbon exchanges, and therefore of poorly predicting future  
3 carbon cycle feedbacks.

4  
5 There is at the same time an urgent need to improve process representation and expand the suite of processes  
6 in the models, including forest regrowth and recovery effects (Sitch et al., 2005), ecosystem demography  
7 (e.g. Moorcroft), carbon-nitrogen interactions (e.g., Nadelhofer et al., 2004) and the effects of temperature-  
8 precipitation covariance (Bonfils et al., 2005; Angert et al., 2005; Hutyra et al., 2005) on the land, and more  
9 complete treatments of ocean ecosystems (e.g., resolving more than one phytoplankton functional type),  
10 including iron and other micronutrient limitations (see 7.3.2.2).

11  
12 [START OF QUESTION 7.1]

13  
14 **Question 7.1: Are the Increases in Atmospheric Carbon Dioxide and Other Greenhouse Gases During**  
15 **the Industrial Era Caused by Human Activities?**

16  
17 *Yes, the increases in atmospheric carbon dioxide and other greenhouse gases in the industrial era are*  
18 *caused by human activities. For carbon dioxide, human activity, including burning fossil fuels and land use*  
19 *change, have released more carbon dioxide to the atmosphere than can be accounted for by greater*  
20 *atmospheric carbon dioxide concentrations. For methane, another important greenhouse gas, human*  
21 *activities contributed almost half of the annual emissions over the last 20 years. For nitrous oxide, human*  
22 *activity contributes about one third to one half of the total nitrous oxide emitted to the atmosphere each*  
23 *year. Most of the halogen containing gases are manufactured by humans, and were not present in the*  
24 *atmosphere before the industrial era.*

25  
26 Human activities are an important, and often dominant source, of the increase in atmospheric concentration  
27 of carbon dioxide and greenhouse gases.

28  
29 *Carbon Dioxide (Panel a)*

30 Fossil fuel combustion, together with cement manufacture, has released about three quarters of the total  
31 carbon dioxide released to atmosphere during the last twenty years. The remainder of the carbon dioxide  
32 emitted to the atmosphere is by land use change, much of which occurs in the tropics.

33  
34 The total quantity of carbon dioxide is exchanged between the land and atmosphere each year is estimated to  
35 be 120 Pg C and between the ocean and atmosphere is estimated to be 90 Pg C. These numbers are generally  
36 in balance, or at steady state for the global carbon cycle, and thus are not the cause of the rise in atmospheric  
37 carbon dioxide. The increase in atmospheric concentration is about 1.5% of the total exchange. A variety of  
38 techniques, including the oxygen to nitrogen ratio, use of the isotopes of carbon for tracers, and model  
39 estimates have been used to determine that human activity is causing the increase in atmospheric carbon  
40 dioxide. These techniques all provide further evidence and support for the partitioning of human driven  
41 emissions, into land, ocean and atmospheric reservoirs as shown in Panel a.

42  
43 [INSERT QUESTION 7.1, FIGURE 1 HERE]

44  
45 *Halogen-Containing Gases (Question 7.1, Figure 1, Panel b)*

46 The halogen-containing gases include chlorofluorocarbons, CFCs, hydrochlorofluorocarbons (HCFCs),  
47 hydrofluorocarbons (HFCs) and halons. Before industrialization, there were only a few naturally occurring  
48 halogen-containing gases, e.g., methyl bromide and methyl chloride. The atmospheric abundances of key  
49 halogen-containing gases produced by humans are shown in Panel b. The development of new techniques for  
50 chemical synthesis resulted in a proliferation of chemically manufactured halogen containing gases during  
51 the last 50 years of the 20th century. As a result of the Montreal Protocol, and its adjustments and  
52 amendments, the concentration of many halogen-containing species, including CFCs is no longer increasing  
53 at the Earth's surface. The abundance of gases not covered by the Montreal Protocol, HCFCs, continues to  
54 increase.

55  
56 *Methane (Question 7.1, Figure 1, Panel c)*

1 Human activities are responsible for almost half of the methane emitted to the atmosphere. The natural  
2 sources of methane to the atmosphere include wetlands, termites, oceans, and methane hydrates. The human  
3 activities that produce methane include energy production, land fills, raising ruminant animals, rice  
4 agriculture and biomass burning. The sinks for methane include chemical oxidation in the troposphere, and  
5 the much smaller soil uptake and eventual destruction in the stratosphere. The atmospheric increase and the  
6 atmospheric abundance of methane are determined by the balance between emissions and destruction.

7  
8 *Nitrous Oxide (Question 7.1, Figure 1, Panel d)*

9 Human activities are responsible for about one-third to one-half of the total emissions of nitrous oxide. The  
10 natural sources of nitrous oxide include oceans, chemical oxidation of ammonia in the atmosphere, and soils.  
11 Tropical soils are a particularly important source of nitrous oxide to the atmosphere. The human activities  
12 that emit nitrous oxide include transformation of fertilizer N into N<sub>2</sub>O and its subsequent emission from  
13 agricultural soils, biomass burning, emissions from cattle and feedlots, and some industrial sources,  
14 including nylon manufacture. Most of the nitrous oxide emitted to the atmosphere is eventually removed by  
15 destruction in the stratosphere. The atmospheric increase and the atmospheric abundance of nitrous oxide are  
16 determined by the balance between emissions and destruction.

17  
18 [END OF QUESTION 7.1]

## 19 20 **7.4 Reactive Gases and the Climate System**

21  
22 The atmospheric concentration of many reactive gases has increased substantially during the industrial era as  
23 a result of human activities. Some of these compounds (methane, nitrous oxide, halocarbons, ozone, etc.)  
24 interact with longwave (infrared) solar radiation and, as a result, contribute to 'greenhouse warming'. Ozone  
25 also absorbs efficiently shortwave (ultraviolet and visible) solar energy, so that it protects the biosphere  
26 (including humans) from harmful radiation and plays a key role for the energy budget of the middle  
27 atmosphere. Many atmospheric chemical species are emitted at the surface as a result of biological processes  
28 (soils, vegetation, oceans) or anthropogenic activities (fossil fuel consumption, land-use changes) before  
29 being photochemically destroyed in the atmosphere, and converted to compounds that are eventually  
30 removed by wet and dry deposition. The oxidizing power (or capacity) of the atmosphere is determined  
31 primarily by the atmospheric concentration of the hydroxyl (OH) radical (daytime) and to a lesser extent of  
32 NO<sub>3</sub> (night time), ozone and H<sub>2</sub>O<sub>2</sub>. The coupling between chemical processes in the atmosphere and the  
33 climate system are complex because they involve a large number of physical, chemical and biological  
34 processes that are not always very well quantified. An important issue is to determine to what extent  
35 predicted climate change could affect air quality. The goal of this Section is assess recent progress made in  
36 the understanding of the two-way interactions between reactive gases and the climate system.

### 37 38 **7.4.1 Methane (CH<sub>4</sub>)**

#### 39 40 **7.4.1.1 Biogeochemistry and present-day budgets of CH<sub>4</sub>**

41 Atmospheric CH<sub>4</sub> originates from both non-biogenic and biogenic sources. Non-biogenic CH<sub>4</sub> includes  
42 emissions from fossil fuel (natural gas, petroleum and coal) burning, biomass burning, and geological  
43 sources (fossil CH<sub>4</sub> of geothermal and volcanic origin). However, emissions from biogenic sources account  
44 for more than 70% of the global total. These sources include wetlands, rice agriculture, livestock, landfill  
45 and termites. CH<sub>4</sub> emission from these sources is an ecosystem process that results from a complex sequence  
46 of events beginning with primary fermentation of organic macromolecules to acetic acid (CH<sub>3</sub>COOH), other  
47 carboxylic acids, alcohols, CO<sub>2</sub> and H<sub>2</sub>, followed by secondary fermentation of the alcohols and carboxylic  
48 acids to acetate, H<sub>2</sub> and CO<sub>2</sub>, which are the ultimate precursors of CH<sub>4</sub> (CH<sub>3</sub>COOH → CH<sub>4</sub>+CO<sub>2</sub> and  
49 CO<sub>2</sub>+4H<sub>2</sub> → CH<sub>4</sub>+2H<sub>2</sub>O). Methane-producing archaea (methanogens) are the only known organisms capable  
50 of these substrate conversions. CH<sub>4</sub> production from these sources is generally limited by availability of  
51 substrates and anaerobiosis conditions. Not all of the CH<sub>4</sub> produced is emitted to the atmosphere. Most of it  
52 (50-90%) is oxidized (i.e. in the presence of molecular oxygen) before emission to the atmosphere, by  
53 methanotrophs or methane oxidizing bacteria, which convert CH<sub>4</sub> into CO<sub>2</sub>, water and microbial biomass.  
54 CH<sub>4</sub> consumption in this case is limited by both O<sub>2</sub> and CH<sub>4</sub> availability and regulated largely by  
55 meteorological variables such as moisture and temperature. Although anaerobic CH<sub>4</sub> oxidation (no molecular  
56 O<sub>2</sub> present) exists in some environments (Orphan et al., 2002; Hallam et al., 2004), its role in contributing to  
57 the global CH<sub>4</sub> budget is highly uncertain. The net rate of CH<sub>4</sub> emissions from sources depends on an

1 imbalance between production and oxidation, which is strongly influenced by climatic and edaphic factors  
2 (temperature, water table, substrates and pH (Conrad, 1996)).

3  
4 Estimating emissions from CH<sub>4</sub> sources is based on three approaches: extrapolation from on-site direct flux  
5 measurements, a process-based modelling approach that represents the actual physical and biological  
6 processes of CH<sub>4</sub> production and emission (bottom-up approach), and inverse modelling that relies on  
7 observations in the atmosphere (top-down approach). Direct flux measurements provide the initial estimates  
8 of upper and lower emission ranges, under the assumption that point measurements represent all locations  
9 that share similar characteristics such as soil or ecosystem type, which is not necessarily true. Therefore,  
10 extrapolation of direct flux measurements to larger scales contains considerable uncertainty. The process-  
11 based modelling approach requires sufficient understanding of local parameters and processes, and assumes  
12 that such characteristics apply to other sites (e.g., Cao et al., 1998; Walter et al., 2000, 2001a, 2001b).  
13 Currently, only a few such locations exist where parameters are investigated to sufficient detail for  
14 modelling. Extrapolation from results of a few sites to regional or global scales thus is still associated with  
15 uncertainties. On the other hand, the top-down approach helps overcome the weak points in bottom-up  
16 methods by bypassing limitations in our understanding of methanogenic-methanotrophic and their  
17 interactions with environmental factors. Inadequate observations and uncertainties in modelled circulation  
18 patterns are the main obstacles for its extensive application (Chen and Prinn, 2005a; Dentener et al., 2003;  
19 Mikaloff Fletcher et al., 2004a, 2004b). Measurements of the isotopes of CH<sub>4</sub> (<sup>13</sup>C and <sup>14</sup>C) can provide  
20 additional constraints on CH<sub>4</sub> budgets and specific sources. However, such data are even more limited  
21 (Mikaloff Fletcher et al., 2004a, 2004b; Lassey et al., 2000).

22 **Table 7.4.1.** Sources, sinks, and atmospheric budgets of CH<sub>4</sub> (Tg-CH<sub>4</sub> yr<sup>-1</sup>)

Reference	Base Year	Methane Sources							Total
		Biogenic Sources					Non-Biogenic Sources		
		Wetlands	Rice Agriculture	Livestock	Landfill & Waste	Termite	Biomass Burning	Energy	
Wuebbles and Hayhoe, 2002	-	123 <sup>a</sup>	60	81	61	20	52 <sup>b</sup>	106	503
Bogner and Matthews, 2003	6	-	-	-	16–57	-	-	-	-
Mikaloff Fletcher et al., 2004a, b	199	221	54	91	35	29	88	82 <sup>c</sup>	610
Wang et al., 2004	4	176	57	83	49	20	41	81 <sup>c</sup>	507
Chen and Prinn, 2005 <sup>d</sup>	199	145	112	189 <sup>e</sup>	-	23	43	84	596
TAR	8	-	-	-	-	-	-	-	598
		Methane Sinks							
		Oxidation by OH in Troposphere		Oxidation in Terrestrial Ecosystem		Loss to Stratosphere			
Wuebbles and Hayhoe, 2002	-	445		30		40			515
Wang et al., 2004	199	428		-		30			492
Ridgewell et al., 1999	-	-		38		-			-
TAR	199	506		30		40			576
	8								

25 Notes:

- 1 (a) Comprises of emission from natural wetland of 100 Tg, oceans 4 Tg, marine sediments 5 Tg and geological sources  
 2 14 Tg  
 3 (b) Includes wild fires emission of 2 Tg  
 4 (c) Includes hydrate emission of 4 Tg  
 5 (d) Inverse result of control scenario in Chen and Prinn, 2005.  
 6 (e) Includes emission from landfill and waste  
 7  
 8

9 Partly due to differences in methodologies used and the nature of data employed, there are currently large  
 10 uncertainties in CH<sub>4</sub> emission estimates of individual sources and how they have varied over the past. The  
 11 currently available range of total annual emissions is 500 to 600 Tg-CH<sub>4</sub> yr<sup>-1</sup> (Table 7.4.1), similar to those  
 12 given in TAR. Although estimates have not changed much since the time reported in TAR, advances have  
 13 been made in constraining individual source strengths. A recent estimate, which optimized emission sources  
 14 based on the previous bottom-up and top-down reported values in combination with the worldwide  
 15 observation results in a 3-D atmospheric transport and chemical model (ATCM) simulation, gives global  
 16 emissions of 596 Tg-CH<sub>4</sub> yr<sup>-1</sup> (Chen and Prinn, 2005a, 2005b). This estimate has significantly reduced the  
 17 uncertainty in global source strength through employing high quality datasets and rigorous inter-laboratory  
 18 calibrations (from 13 high-frequency stations that measured methane concentration between 24 and 36 times  
 19 per day, and flask samples collected periodically between 1966 and 2001 (once per week) from locations  
 20 situated worldwide). Uncertainty in estimates of northern wetland emission has been greatly reduced (<25%  
 21 of means) due to the existence of numerous high-frequency monitoring stations. Results from this study  
 22 indicate that southern and tropical regions account for >70% of total global wetland emissions. During 1996-  
 23 2001, significantly reduced emissions are suggested from energy use (coal and natural gas) and slightly for  
 24 wetlands while significantly increased emissions are found from animals and wastes, wetland rice agriculture  
 25 (20% increase, mostly between 0-30°N) and tropical biomass burning (30-80% since 1996). In other studies  
 26 that included both observation results and <sup>13</sup>C/<sup>12</sup>C ratios of CH<sub>4</sub>, an increase in emissions from tropical  
 27 wetlands is also suggested (Mikaloff Fletcher et al., 2004a; Wang et al., 2004). Such increased emissions  
 28 probably result from temporary wetlands that are flooded for only part of the year (during the rainy season,  
 29 Kaplan, 2001), and thus are not accounted for in bottom-up estimates. Recently, advances have been also  
 30 made on mapping global distribution of methane by using space-born near-infrared absorption spectroscopy  
 31 (Frankenberg et al., 2005). The results from such observations suggest high emission from tropical  
 32 rainforest, supporting the findings from top-down inversion results of Mikaloff Fletcher et al. (2004a) and  
 33 Chen and Prinn (2005). It seems, therefore, that the reduced emissions, notably in energy use and northern  
 34 wetland lands, are compensated by increased emission from lower latitudes. Such offsetting changes in  
 35 sources and sinks may be one of the reasons for the temporary slow down in the current growth rate (Chapter  
 36 2). However, an increase in CH<sub>4</sub> emissions from certain wetlands was observed in 2001 (Table 7.4.2),  
 37 suggesting that the current slowdown of CH<sub>4</sub> growth may be temporary (Chen and Prinn, 2005a).  
 38

39 **Table. 7.4.2.** Control Case annual anomalies of emission trend during 1996–2001 compared to the deviation  
 40 from mean during the same period. The values are expressed in Tg-CH<sub>4</sub> yr<sup>-1</sup> (Chen and Prinn, 2005b).  
 41

Region	1996	1997	1998	1999	2000	2001
Wetland-Northeast <sup>a</sup>	2.7	1.9	1.5	-2.8	-2.0	2.5
Wetland-Northwest <sup>b</sup>	-1.9	1.5	2.2	0.2	-1.6	-0.4
Wetland-South <sup>c</sup>	2.5	3.2	5.3	-3.2	-10.7	2.8
Rice agricultured	-7.5	-4.8	22.3	-1.4	1.9	-10.4
Biomass burning (Africa)	0.8	1.1	0.5	0.5	-0.6	-2.3
Biomass burning (America)	1.6	1.4	0.5	0.5	-3.7	-0.4
Biomass burning (Asia)	-0.1	-1.2	0.8	-0.3	0.9	-0.2
Wetland Total	1.7	1.8	13.7	-6.1	-13.8	2.7
Rice Total	-5.9	-3.8	17.6	-1.1	1.5	-8.2
Biomass Total	2.3	1.3	1.8	0.8	-3.4	-2.8
Overall Total	-1.9	-0.7	33.1	-6.5	-15.7	-8.3

42 Notes:

43 (a) North America and Canada

44 (b) Europe and Russia

45 (c) Below 30°S

46 (d) 21% of area is overlapped with wetlands

1  
2  
3 Results from both a process-based model for 1984–1999 (Dlugokencky et al., 2001) and an inverse model  
4 for 1996–2001 (Chen and Prinn, 2005) indicate that the largest contributors to global inter-annual variation  
5 in emissions during 1996–2001 are emissions from rice agriculture and wetlands. Contributions from  
6 biomass burning are relatively small. However, errors may arise in estimates of biomass burning due to lack  
7 of data (mainly undersampling of the total amount of burning). CH<sub>4</sub> emissions from biomass burning also  
8 depend on fire type, which could be highly variable resulting in a large uncertainty.

9  
10 Wetland emissions were suggested to account for the large increase in emissions in 1998 (Dlugokencky et  
11 al., 2001; Mikaloff Fletcher et al., 2004; Chen and Prinn, 2005b). The ENSO event that occurred in late 1997  
12 and early 1998 associated with enhanced precipitation at low latitudes likely stimulated methanogenesis in  
13 low latitude wetlands (Bell et al., 1999). Chen and Prinn (2005) partitioned this wetland contribution into  
14 northern wetlands (5–12 Tg), tropical wetlands (8–13 Tg) and rice agriculture (10 Tg) (Table 7.4.2).  
15 Unusually warm and dry conditions in the northern hemisphere during the ENSO period also increased  
16 biomass burning. CH<sub>4</sub> releases of 3–5 Tg in 1998 were attributed to boreal forest fires in Eastern Siberia due  
17 to these unusual warm and dry conditions (Kasischke et al., 2002).

18  
19 For rice agriculture, there has been a downward trend over the past few decades in estimates of CH<sub>4</sub>  
20 emissions using bottom-up methods. This trend is largely due to results from the increasing number of in-  
21 situ flux measurements used for emission extrapolation, which collectively indicate lower overall emissions  
22 from rice agriculture. However, emission estimates still vary greatly in both space and time. The non-  
23 homogeneous nature of paddy-field soils and different cultivation practices mainly account for such  
24 variations (Aulakh et al., 2001; Li et al., 2002). Sass et al. (2002) measured emissions at a single site in the  
25 U.S. over nine years and observed a year-to-year flux variability of approximately ±50% of the annual mean  
26 over the entire period. Using the standard deviation of mean CH<sub>4</sub> flux as a measure of variability, they found  
27 that accounting for rice plant height and grain yield reduced spatial variability from 25.2 to 17.7% of the  
28 mean. Temporal variability over the entire 9-year data set was 49% of the mean, 71% of which was  
29 explained by variations in average rice plant height and total nitrogen fertilizer application. This highlights  
30 the finding that even with a consistent measurement approach, there remains a year-to-year variation in  
31 emissions, and that emissions are highly sensitive to local factors. Global estimates of emissions from this  
32 source will still likely contain a certain level of variation due to such factors.

33  
34 For landfills, estimates of CH<sub>4</sub> emissions are normally based on measured or estimated national solid waste  
35 generation, the fraction landfilled, the fraction landfilled that is expected to biodegrade anaerobically, the  
36 degradable organic carbon content of that fraction, and the CH<sub>4</sub> content of the biogas product. For some  
37 developed countries, subtractions are made for methanotrophic CH<sub>4</sub> oxidation in cover soils and CH<sub>4</sub>  
38 recovery via pumped system. In general, estimates still contain greater uncertainty when compared with  
39 emissions from wetland and rice agriculture, since solid waste data are lacking for many countries, reliability  
40 of existing data is questionable, no internal variability is available, there is a wide range in magnitude (7  
41 order difference, Bogner and Mathews, 2003), and there are no international networks/campaigns to measure  
42 landfill CH<sub>4</sub> emissions. Bogner and Mathews (2003) developed a methodology for estimating CH<sub>4</sub> emission  
43 from landfills during 1980–1996, based on per capita energy generation to estimate the amount of solid  
44 waste generation. Their best estimate ranged between 16 and 57 Tg, which is within the ranges reported for  
45 other studies (e.g., in TAR).

46  
47 Once in the atmosphere, CH<sub>4</sub> is destroyed through two main mechanisms: oxidation by OH in the  
48 troposphere and by methanotrophs in terrestrial ecosystems. Some CH<sub>4</sub> is also lost to the stratosphere.  
49 Current estimates for these sinks combined span 492–576 Tg-CH<sub>4</sub> yr<sup>-1</sup>. The abundance of OH (Section 7.4.2)  
50 and its chemistry determine CH<sub>4</sub> sink strength, the lifetime of CH<sub>4</sub> and its growth rate in the atmosphere.  
51 Current observations indicate a stabilized growth rate (Chapter 2) which may result from changes in such  
52 OH chemistry. However, observed trends in the seasonal cycle of CH<sub>4</sub> and OH concentrations inferred from  
53 methylchloroform measurements (a strong positive trend of 15±22% during 1979–1989, followed by a  
54 strong decrease the following decade, Prinn et al., 2001) do not support a continued decrease in CH<sub>4</sub> growth  
55 rate. It appears, then, that changes in emissions rather than increasing sink strengths are responsible for the  
56 major part of the observed slowing.

1 Consumption of atmospheric CH<sub>4</sub> in terrestrial ecosystem ranges between 20 and 51 Tg-CH<sub>4</sub> yr<sup>-1</sup>, with the  
2 best estimates around 30–38 Tg (TAR, 1998; Ridgwell et al., 1999). Over recent decades, this sink strength  
3 has probably decreased due to intensification of agriculture that alters the methanotrophic community and  
4 activity (Knief et al., 2005). Changes in land use, especially cultivation of formerly undisturbed soils, reduce  
5 the sink strength for atmospheric methane by 60–90% (Whalen and Reeburgh, 1990; Steinkamp et al., 2001;  
6 Knief et al., 2005). The impact is most pronounced in the dry tropical forest where sink strength accounts for  
7 a third of the total global sink and where rapid deforestation is occurring (Keller and Reiners, 1994). An  
8 estimate for the effect of land use change and agricultural intensification is a 3 Tg yr<sup>-1</sup> reduction in sink  
9 strength (Ridgwell et al., 1999).

10  
11 Assuming a combined sink of ~576 and a combined source of ~596 Tg-CH<sub>4</sub> yr<sup>-1</sup> yields an imbalance of 20  
12 Tg-CH<sub>4</sub> yr<sup>-1</sup> in source emissions over sink removals.

#### 13 14 7.4.1.2 *Effects of climate*

15 We learn about the effects of climate on CH<sub>4</sub> biogeochemistry mainly from examining records of the past  
16 and from model simulations under various climate change scenarios. The Vostock ice core record back to  
17 420,000 years BP (Petit et al., 1999) reveals that atmospheric concentration is closely tied to atmospheric  
18 temperature records, decreasing and rising in phase with temperature at the inception and termination of  
19 glacial episodes (Wuebbles and Hayhoe, 2002). Another study showed that following each transition,  
20 temperature increased more rapidly than CH<sub>4</sub> concentration (Brook et al., 2000). Since biogenic CH<sub>4</sub>  
21 production and emission from major sources (wetland, landfill, rice agriculture) are temperature-dependent,  
22 climate change scenarios with a warmer atmosphere, suggest enhanced emissions from these sources.

23  
24 Wetlands are the largest CH<sub>4</sub> emitter globally (25–40% of total global emission, Table 7.4.1). Several studies  
25 have demonstrated a high sensitivity of wetlands CH<sub>4</sub> emission to temperature and water table. Before the  
26 1990s global CH<sub>4</sub> emissions increased steadily, due both to an increase in generic sources, and to elevated  
27 surface temperature leading to an increased emission from wetlands (Walter et al., 2001a, 2001b;  
28 Christensen et al., 2003; Zhuang et al., 2004). The sensitivity of wetland emissions to climate varies  
29 depending on regional and local factors. Earlier estimates of the effect of warming on CH<sub>4</sub> emission were  
30 given by Chapman and Thurlow (1996). From the relationship between emission and temperature at two  
31 sites in Scotland, they predicted that CH<sub>4</sub> emission would increase by 17, 30 and 60% for warmings of 1.5,  
32 2.5 and 4.5 °C (warming above the site's mean temperature during 1951–1980). A model simulation by Cao  
33 et al. (1998) yielded a 19% emission increase under a uniform 2°C warming. The combined effects of 2°C  
34 warming and 10% increase in precipitation yielded an increase of 21% in emissions. In most cases, the net  
35 emission depends on how an increase in temperature affects net ecosystem production (NEP), as this is the  
36 source of methanogenic substrates (Christensen et al., 2003), and on the moisture regime of wetlands, which  
37 determines its aerobiosis/anaerobiosis. Emissions are enhanced under a scenario where an increase in  
38 temperature is associated with increases in precipitation and NEP. On the other hand, emission decreases if  
39 elevated temperature results in either reduced precipitation or NEP.

40  
41 Walter and Heimann (2000) assessed how changes in water table and temperature would affect emission in  
42 several northern wetlands. When the water table is above the soil surface, CH<sub>4</sub> emission follows soil  
43 temperature. If the water table is below the soil surface, the pattern of emission is mainly driven by changes  
44 in soil temperature but the amplitude is influenced by the water table depth. An increase in temperature of  
45 1°C results in 20% increase in simulated CH<sub>4</sub> emission while a water table increase of 10 cm would increase  
46 emission 0–25%, depending on initial wetness and dryness of wetlands. Under dry conditions, an increase in  
47 the water table enhances CH<sub>4</sub> emission significantly while under wet conditions water table changes have  
48 only a small effect.

49  
50 A standard benchmark for climate change studies is a doubling in CO<sub>2</sub> concentration. Shindell et al. (2004)  
51 have applied a GCM to simulate a 3.4°C due to this doubling in CO<sub>2</sub> and found that CH<sub>4</sub> emission increases  
52 by 78% above current emissions from wetlands. Zhuang et al. (2004) use a terrestrial ecosystem model  
53 (TCM) to study how rates of CH<sub>4</sub> emission and consumption in high-latitude soils of the Northern  
54 Hemisphere (north of 45°N) have changed over the past century (1900–2000) in response to observed change  
55 in the region's climate, based on the emission data for the 1990s. They estimate that net emissions of CH<sub>4</sub>  
56 have increased by an average 0.08 Tg-CH<sub>4</sub> yr<sup>-1</sup> during 20th century. The decadal net CH<sub>4</sub> emission rate  
57 correlates with soil temperature, water table depth and NPP. Estimates of emission from northern wetlands

1 also depend largely on the formulation of the active layer (for methanogenesis): increasing the active layer a  
2 mere 10 cm could increase net CH<sub>4</sub> emission as much as 38% (Zhuang et al., 2004). In summary, although  
3 sensitivity to climate change differs among wetlands, most studies point towards increasing emission trends  
4 under warmer temperature and enhanced precipitation scenarios.

5  
6 In rice agriculture, climate factors that will likely influence CH<sub>4</sub> emission are those associated with plant  
7 growth. Plant growth determines how much substrate will be available for either methanogenesis or  
8 methanotrophy and thus the net emission (Matthews and Wassmann, 2003). As mentioned above, plant  
9 growth (height) strongly correlates with CH<sub>4</sub> emission in a Texas rice field (Sass et al., 2002). Any climate  
10 change scenario that results in an increase in plant biomass is likely to increase CH<sub>4</sub> emission from rice  
11 agriculture (Xu et al., 2004). However, the magnitude of increased emission depends largely on field  
12 management. For instance, field drainage could significantly reduce emission due to the introduction of  
13 aerobiosis in the soil (i.e. influx of air into anaerobic zone which subsequently suppresses methanogenesis).

14  
15 Climate also affects CH<sub>4</sub> sinks. Several model studies indicate that CH<sub>4</sub> oxidation in soil is relatively  
16 insensitive to temperature increase (Ridgwell et al., 1999; Zhuang et al., 2004). A doubling of CO<sub>2</sub> would  
17 likely change the sink strength only marginally (in the range of -1 to +3 Tg yr<sup>-1</sup>, Ridgwell et al., 1999).  
18 However, any change in climate that leads to altering the amount and pattern of precipitation may  
19 significantly affect the CH<sub>4</sub> oxidation capacity of soils. A process-based model simulation indicated that CH<sub>4</sub>  
20 oxidation strongly depends on soil gas diffusivity, which is a function of soil bulk density and field capacity  
21 (Bogner et al., 2000; Del Grosso et al., 2000).

22  
23 In summary, since TAR advances have been made in constraining estimates of CH<sub>4</sub> source strengths and in  
24 understanding of emission variations. These improvements are attributed mainly to increasing availability of  
25 measurement data worldwide and improved modelling tools (e.g., Chen and Prinn 2005a, 2005b; Mikaloff  
26 Fletcher, 2004a, 2004b). An understanding of climate effects on CH<sub>4</sub> sinks and sources has evolved, and  
27 such effects have been quantified. As a result, wetlands and rice agriculture are identified as the CH<sub>4</sub> sources  
28 that are most sensitive to climate change and thus will likely contribute most to changes in global CH<sub>4</sub>  
29 budgets in the future. A warm and wet climate would likely enhance emission from these sources while a  
30 warm but dry climate would likely reduce emission. Since these sources are located in the tropics where  
31 measurements are lacking, uncertainty in future emission estimates could be reduced by an increase in  
32 measurement coverage in such regions.

#### 33 34 **7.4.2. Nitrogen Compounds**

35  
36 The nitrogen cycle is important to the functioning of the earth system and to climate (Figure 7.4.1, Holland  
37 et al., 2005). Nitrogen is a major limiting nutrient in terrestrial and aquatic ecosystems and an important  
38 catalyst in tropospheric photochemistry. Over the last century human activities have dramatically increased  
39 inputs of reactive nitrogen to the global atmosphere by as much as three to five fold. Reactive nitrogen is the  
40 combination of oxidized, reduced and organically bound nitrogen but not the atmospherically abundant N<sub>2</sub>  
41 gas. The impact of the change in the N cycle has led to problems such as compromised air quality and human  
42 health, acidification of ecosystems, degradation of coastal estuaries, and acceleration of the eutrophication of  
43 lakes (Vitousek et al., 1997; Rabalais, 2002; Townsend et al., 2003; Galloway et al., 2004).

44  
45 [INSERT FIGURE 7.4.1 HERE]

46  
47 Perturbations of the nitrogen cycle impact the atmosphere climate system through production of three key N  
48 containing trace gases: nitrous oxide (N<sub>2</sub>O), ammonia (NH<sub>3</sub>) and nitrogen oxides (NO<sub>x</sub>=NO+NO<sub>2</sub>).

##### 49 50 **7.4.2.1 Nitrous Oxide (N<sub>2</sub>O), Nitrogen Oxides (NO<sub>x</sub>=NO+NO<sub>2</sub>), and Ammonia (NH<sub>3</sub>)**

51 Nitrous oxide is the nitrogen trace gas with the longest atmospheric lifetime, 120 years (range 97–137)  
52 (Prather et al., 1998). Nitrous oxide concentrations serve as a tracer for human manipulation of the global  
53 nitrogen cycle. The exponential rise in tropospheric N<sub>2</sub>O concentrations shown in Figure 7.4.2.a confirms the  
54 profound modification of the global nitrogen cycle. Tropospheric abundances of N<sub>2</sub>O have increased from  
55 pre-industrial values of about 270 ppb (TAR) to between 315 and 319 ppbv (CMDL, ALE-GAGE).  
56 Estimates of the global N<sub>2</sub>O budgets have changed little since the TAR. The sources and sinks remain out of  
57 balance, thus generating a continuing increase in atmospheric concentrations of N<sub>2</sub>O. Much of the imbalance

globally can be attributed to the intensification of human activity, particularly agriculture, (Galloway et al. 2005). The average annual growth rate for 1999–2000 was 0.85–1.1 ppbv yr<sup>-1</sup> (WMO Ozone Assessment 2002). The emissions of N<sub>2</sub>O from soils, natural plus agricultural, remain the dominant source of N<sub>2</sub>O to the atmosphere (Bouwman et al., 2002b, c). Since the TAR, much of the focus has been on refining the estimates of N<sub>2</sub>O from agricultural sources, the single biggest anthropogenic source of N<sub>2</sub>O (Del Grosso, 2005; Smith and Conen, 2004). The long lifetime of atmospheric N<sub>2</sub>O also means that it plays an important role in stratospheric ozone depletion. Nitrous oxide is likely to become increasingly important as the emissions of atmospheric halogens decline in compliance with the Montreal Protocol.

[INSERT FIGURE 7.4.2 HERE]

Measurements of changes in atmospheric concentrations of NO<sub>x</sub> and NH<sub>x</sub> (NH<sub>3</sub> + NH<sub>4</sub><sup>+</sup>) are challenging to establish because the atmospheric lifetimes of these chemical species are days to weeks instead of years. The short atmospheric lifetimes of NO<sub>x</sub> and NH<sub>x</sub> generate pronounced spatial and temporal variations in their distribution. Their atmospheric concentrations are much more variable regionally and through time than are measurements of N<sub>2</sub>O. Figure 7.4.2a depicts the exponential increase in fossil fuel NO<sub>x</sub> emissions. Total global NO<sub>x</sub> emissions have increased from an estimated 11 Tg-N y<sup>-1</sup> pre-industrially (Holland et al., 1999; Galloway et al., 2004) to between 35 and 50 Tg-N y<sup>-1</sup> for 2000 and are forecast to be 105–131 Tg-N y<sup>-1</sup> by 2100 (Lamarque et al., 2005). The estimates of contemporary and future NO<sub>x</sub> emissions were based on 29 simulations by 6 tropospheric chemistry models using the A2 scenario projections for 2100 (Lamarque et al., 2005). Land use change continues to have a significant impact on both NO and N<sub>2</sub>O emissions. Land use change impacts NO: recent estimates show that logging increases N<sub>2</sub>O and NO emissions by 30–350% depending on conditions. Interactions between soil emissions and scavenging by plant canopies have a significant impact on soil NO<sub>x</sub> emissions to the free troposphere; the impact may be greatest in regions where fossil fuel emissions are rising, in subtropical and tropical regions (Ganzeveld et al., 2002).

Since pre-industrial times, there has been an exponential increase in all the available indices of the intensity of agricultural nitrogen cycling, the primary source for NH<sub>3</sub> emissions (Bouwman et al., 2002a, b & c; Figure 7.4.2b and Table 7.4.3). Total global NH<sub>3</sub> emissions have increased from an estimated 11 Tg-N y<sup>-1</sup> pre-industrially to 54 Tg-N y<sup>-1</sup> for 2000, and are projected to increase to 116 Tg-N y<sup>-1</sup> for 2050 (Holland et al., 1999; Galloway et al., 2004).

**Table 7.4.3.** Global sources (in Tg-N yr<sup>-1</sup>) of NO<sub>x</sub>, NH<sub>3</sub>, and N<sub>2</sub>O. (From Bouwman et al., 2002a, b, c).

Source	NO <sub>x</sub>	NH <sub>3</sub>	N <sub>2</sub> O
Anthropogenic sources			
Fossil fuel	21.9	0.1	0.2
Industrial processes	1.7	0.2	0.3
Animal husbandry	0.7	21.6	3.4
Fertilizer and crops	0.4	12.6	2.2
Biomass burning	7.7	5.9	0.7
Human excreta	–	2.6	0.2
Coastal water	–	–	1.9
Atmospheric deposition	0.3	–	0.6
Natural sources			
Soils under natural vegetation	13.0	2.4	6.6
Oceans	–	8.2	3.6
Lightning	12.2	–	–
Tropospheric chemistry	0.9	–	0.6
Stratospheric chemistry	0.7	–	–
Total source	59.5	53.6	20.3

Because the atmospheric lifetimes of NO<sub>x</sub> and NH<sub>x</sub> are relatively short, 1 day–2 weeks, quantification of changes in atmospheric concentrations over time are harder to establish than for nitrous oxide or carbon

dioxide. The short atmospheric lifetime also means that the deposition of  $\text{NH}_x$  and  $\text{NO}_x$  and their reactions products, including aerosols, is the primary mechanism for removing these chemical species from the atmosphere. Estimates of the rate of removal of both  $\text{NH}_x$  and  $\text{NO}_x$  are provided by measurements of atmospheric deposition that have been conducted over the US and Western Europe to quantify acid rain inputs (Holland et al., 2004). Evaluation of these data for the 25 years of available measurements suggests that there is substantial seasonal and interannual variability in the wet deposition of  $\text{NH}_x$  and of  $\text{NO}_3$ —the primary removal product(s) for  $\text{NH}_3$  and  $\text{NO}_x$  emissions respectively. However, no clear trend in the deposition of either compound, or in precipitation, emerges over the last 25 years. Chemical transport models represent the deposition removal of  $\text{NO}_x$  and  $\text{NO}_x$  reaction products and  $\text{NH}_x$  and  $\text{NH}_x$  reaction products. A recent intercomparison of 29 simulations and 6 different tropospheric chemistry models focusing on present-day and 2100 conditions for  $\text{NO}_x$  and its removal products forecasts that average N deposition over land will increase by a factor of 2.5 in 2100, mostly due to increases in  $\text{NO}_x$  emissions (Lamarque et al., 2005). Climate contributions to the changes in oxidized N deposition are limited. In Brazil, changes in land use can change the amount of N deposited by as much as fourfold (Lara et al., 2005).

#### 7.4.2.2. Carbon nitrogen interactions

Estimated and projected terrestrial carbon storage requires nutrient inputs, especially nitrogen (Hungate et al., 2003). Nitrogen is a primary limiting nutrient throughout mid and high latitude terrestrial ecosystems, and an important limiting nutrient for plant growth throughout for subtropical and tropical terrestrial ecosystems (Vitousek et al., 1998). Additional nitrogen supply through fertilization and deposition increases plant growth (Vitousek, 2004). When labelled nitrogen ( $^{15}\text{N}$ ) is added to soil and litter layers, much of the additional nitrogen remains there and does not translate into increased carbon storage in wood after 7 years of N addition (Nadelhoffer et al., 2004). Studies of canopy uptake of atmospheric nitrogen suggest that this may be an important mechanism for nitrogen uptake and potentially for carbon storage (Sievering et al., 2000). Nitrogen deposition is spatially correlated with increased atmospheric ozone. Because ozone has a detrimental effect on plant growth, the combined net effect of N deposition and high atmospheric ozone concentrations on ecosystem carbon storage requires further examination (Holland and Carroll, 2003, Ollinger and Aber, 2002). Inclusion of the nitrogen cycle into global carbon cycle models (Figure 7.4.3) can cause a significant reduction in global carbon uptake in response to a 2°C warming because of carbon and nitrogen feedbacks within the plant and microbial system.

[INSERT FIGURE 7.4.3 HERE]

#### 7.4.3. Molecular Hydrogen

Atmospheric  $\text{H}_2$  has recently received increased attention, because of its potential role as an indirect greenhouse gas and the expected perturbations of its budget in a prospective *hydrogen economy* (Tromp et al., 2003; Schultz et al., 2003; Warwick et al., 2004). Potential consequences of drastically increased  $\text{H}_2$  emissions include a reduction of the global oxidizing capacity (presently  $\text{H}_2$  constitutes 5–10% of the global average OH sink, Schultz et al., 2003), and increased formation of water vapour, which could lead to increased cirrus formation in the troposphere and additional cooling in the stratosphere, thereby leading to more efficient ozone depletion (Tromp et al., 2003).

There have been several studies of the global tropospheric  $\text{H}_2$  budget (see Table 7.4.4), and they generally agree on a total source strength between 70 and 90  $\text{TgH}_2\text{ yr}^{-1}$ , which is approximately balanced by a sink of equal magnitude. About half of the  $\text{H}_2$  is produced in the atmosphere via photolysis of formaldehyde ( $\text{CH}_2\text{O}$ ), which itself originates from the oxidation of  $\text{CH}_4$  and other volatile organic compounds. The other half stems mostly from the combustion of fossil fuels (e.g., car exhaust) and biomass burning. About 10% of the global  $\text{H}_2$  source is due to ocean biochemistry and nitrogen fixation in soils. Presently, about 50  $\text{TgH}_2\text{ yr}^{-1}$  are produced in the industrial sector, mostly for use in the petrochemical industry (e.g., refineries) (Lovins, 2003). Evaporative losses from industrial hydrogen are generally assumed to be negligible (Zittel and Altmann, 1996). The dominant sink process of atmospheric  $\text{H}_2$  is deposition with catalytic destruction by soil micro-organisms and possibly enzymes (Conrad and Seiler, 1981). The seasonal cycle of the observed  $\text{H}_2$  concentrations implies an atmospheric lifetime of about 2 years (Novelli et al., 1999; Simmonds et al., 2000; Hauglustaine and Ehhalt, 2002), whereas the lifetime with respect to OH oxidation is 9–10 years. This implies that the deposition sink is about 3 to 4 times as large as the oxidation. Loss of  $\text{H}_2$  to the stratosphere and the subsequent escape to space is negligible for the tropospheric  $\text{H}_2$  budget, because there is no

1 significant gradient between tropospheric and stratospheric H<sub>2</sub> mixing ratios, and the budgets of the  
2 troposphere and stratosphere are largely decoupled (Warneck, 1988).

3  
4 **Table 7.4.4.** Summary of global budget studies of atmospheric H<sub>2</sub> (Tg-H yr<sup>-1</sup>).  
5

	Sanderson et al. (2003a)	Hauglustaine and Ehhalt (2002)	Novelli et al. (1999)	Ehhalt (1999)	Warneck (1988)	Seiler and Conrad (1987)
<i>Sources</i>						
Oxidation of CH <sub>4</sub> and VOC	30.2	31	40 ± 16	35 ± 15	50	40 ± 15
Fossil fuel combustion	20	16	15 ± 10	15 ± 10	17	20 ± 10
Biomass burning	20	13	16 ± 11	16 ± 5	15	20 ± 10
N <sub>2</sub> fixation	4	5	3 ± 1	3 ± 2	3	3 ± 2
Ocean release	4	5	3 ± 2	3 ± 2	4	4 ± 2
Volcanoes	–	–	–	–	0.2	–
<b>Total</b>	78.2	70	77 ± 16	71 ± 20	89	87
<i>Sinks</i>						
Deposition	58.3	55	56 ± 41	40 ± 30	78	90 ± 20
Oxidation by OH	17.1	15	19 ± 5	25 ± 5	11	8 ± 3
<b>Total</b>	74.4	70	75 ± 41	65 ± 30	89	98

6  
7  
8 Estimates of the global amounts of H<sub>2</sub> necessary to fuel a future carbon-free energy system are highly  
9 uncertain and depend on the technological parameters as well as the fraction of energy that would be  
10 provided by hydrogen. In the future, hydrogen emissions could at most double compared to the present, and  
11 the impacts on the global oxidizing capacity and stratospheric temperatures and ozone concentrations are  
12 estimated to be small (Schultz et al., 2003; Warwick et al., 2004). As pointed out by Schultz et al. (2003), the  
13 side effects of a global hydrogen economy could have a much stronger impact on the global climate and air  
14 pollution. The global oxidizing capacity is predominantly controlled by the concentration of NO<sub>x</sub> and the  
15 large-scale introduction of hydrogen powered vehicles would lead to a significant decrease of global NO<sub>x</sub>  
16 emissions, leading to a reduction of OH on the order of 5–10%. Reduced NO<sub>x</sub> levels could also significantly  
17 reduce tropospheric ozone concentrations in urban areas. In spite of the expected large-scale use of natural  
18 gas for H<sub>2</sub> production, the impact of a hydrogen economy on the global CH<sub>4</sub> budget is likely to be small,  
19 except for the feedback between the reduced oxidizing capacity (via the NO<sub>x</sub> reduction) and the CH<sub>4</sub>  
20 lifetime.

## 21 22 **7.4.4 Global Tropospheric Ozone**

### 23 24 **7.4.4.1 Present-day budgets of ozone and its precursors**

25 Tropospheric ozone is (after CO<sub>2</sub> and methane) the third most important contributor to greenhouse radiative  
26 forcing since preindustrial times. Typical concentrations are 10–50 ppbv in the lower troposphere, with  
27 higher values in polluted regions, and 40–100 ppbv in the upper troposphere (Logan, 1999). Long-term  
28 trends in these concentrations over the 20th century are discussed in Chapter 2. Ozone is produced within the  
29 troposphere by photochemical oxidation of CO, methane, and nonmethane volatile organic compounds  
30 (NMVOCs) in the presence of nitrogen oxide radicals (NO<sub>x</sub> = NO + NO<sub>2</sub>). Stratosphere-troposphere  
31 exchange (STE) is another source of ozone to the troposphere. Loss of tropospheric ozone takes place by  
32 chemical reactions and by dry deposition to the surface. Estimating the global chemical production and loss  
33 rates of tropospheric ozone, and the implications for ozone concentrations, requires 3-D tropospheric  
34 chemistry models that describe the complex nonlinear chemistry involved and its coupling to transport.  
35 Evaluation of these models with observations of ozone, its precursors, and related species can then be used to  
36 test and improve our understanding of the budget and of the underlying processes.  
37

The past decade has seen considerable development in capabilities for modeling tropospheric ozone chemistry. These were reviewed by the TAR, which reported global tropospheric ozone budgets from eleven models documented in the 1996–2000 literature. We present in Table 7.4.5 the means and standard deviations from these TAR budgets together with results from a number of more recent models including a recent intercomparison of 25 models (Stevenson et al., 2005). It is well established that chemical production and loss are the principal terms in the global budget. Recent models and measurements have shown that the spring maximum of tropospheric ozone in the northern hemisphere, long thought to be stratospheric origin, is in fact largely driven by in situ chemical production (Wang et al, 1998; Lelieveld and Dentener, 2000; Browell et al., 2003; Liu et al., 2004). Though STE is only a minor term in the global budget, it delivers ozone to the upper troposphere where its lifetime is particularly long and where it is of most importance from a radiative forcing perspective.

[INSERT TABLE 7.4.5 HERE]

The recent models in Table 7.4.5 show three significant differences relative to the older-generation TAR models: a weaker STE source, stronger chemical production and loss, and a larger tropospheric ozone burden. It is now well established that many of the older studies overestimated STE, as robust constraints from observed  $\text{NO}_y\text{-N}_2\text{O-O}_3$  correlations in the lower stratosphere impose an STE ozone flux of  $540 \pm 140 \text{ Tg yr}^{-1}$  (Olsen et al., 2001). Overestimation of the STE flux appears to be most serious in models using assimilated meteorological data, due to the effect of assimilation on vertical and horizontal motions (Douglass et al., 2003; Schoeberl et al., 2003; Tan et al., 2004; Van Noije et al., 2004). Simulations with free-running GCM fields tend to have a better stratospheric residual circulation and cross-tropopause transport (Douglass et al., 2003; Shindell et al., 2003; Hauglustaine et al., 2004; Rotman et al., 2004). Hauglustaine et al. (2004) found an increase in the STE ozone flux from  $523 \text{ Tg yr}^{-1}$  in their free running GCM to  $783 \text{ Tg yr}^{-1}$  in the same GCM with winds relaxed toward assimilated meteorology. The newer models using assimilated meteorological data correct for this effect by using dynamic flux boundary conditions in the tropopause region (McLinden et al., 2000) or by relaxing model results to observed climatology (Horowitz et al., 2003). Such corrections, although matching the global STE flux constraints, may still induce errors in the location of the transport (Hudman et al., 2004) with implications for the degree of stratospheric influence on tropospheric concentrations (Fusco and Logan, 2003).

Explaining the faster chemical production and loss of ozone in the current generation of models is less straightforward. Comparisons of newer vs. older generations of the same models suggests that this could reflect improved treatment of NMVOC sources and chemistry (Houweling et al., 1998), UV actinic fluxes (Bey et al., 2001), and deep convection (Horowitz et al., 2003), as well as higher  $\text{NO}_x$  emissions (Stevenson et al., 2005). These explanations would also account for the higher ozone burdens. Another contributing factor is that as the STE source decreases, chemical production increases to compensate, reflecting the nonlinearity of ozone chemistry. The compensating increase could be larger than the decrease in the STE source because ozone injected by STE has on average a longer lifetime than ozone chemically produced in the troposphere (Wauben et al., 1998; Lamarque et al., 2005).

Detailed budgets of ozone precursors were presented in the TAR. The most important precursors are methane and  $\text{NO}_x$  (Wang et al., 1998; Fiore et al., 2002; Grenfell et al., 2003; Dentener et al., 2004). Methane is in general not simulated explicitly in ozone models and is instead constrained from observations.  $\text{NO}_x$  is explicitly simulated and proper representation of its sources and chemistry is critical for the ozone simulation. Table 7.4.6 gives global  $\text{NO}_x$  source estimates from the TAR, including likely ranges (mainly from Holland et al., 1999) and the values used in the TAR OxComp intercomparison of ozone models. The latter assumed a large global fossil fuel source ( $33 \text{ Tg-N yr}^{-1}$ ) to account for increase of Asian emissions over the past decade, however the Asian increase has been compensated by a European decrease (Naja et al., 2003). Also shown in Table 7.4.6 are the  $\text{NO}_x$  sources used in the more recent model studies of Table 7.4.5.

**Table 7.4.6.** Global emissions of  $\text{NO}_x$  ( $\text{Tg-N yr}^{-1}$ ) in the present-day atmosphere<sup>a</sup>

Reference	Fossil Fuel (surface)	Aircraft	Biomass Burning and Biofuels	Soils	Lightning	Total
TAR <sup>b</sup>	33 (20–24)	0.7 (0.2–0.9)	7 (3–13)	6 (4–21)	5 (3–13)	52 (31–73)
Lelieveld and	24	0.5	9	5	5	46

Dentener (2000)						
Bey et al. (2001) <sup>c</sup>	23	0.5	12 (6)	7 (5)	3 (6)	46 (42)
Horowitz et al. (2003)	23	NR	12	7	3	45
Shindell et al. (2003)	21	0.6	6	6	7	41
Von Kuhlmann et al. (2003)	24	0.5	8	6	5	43
Hauglustaine et al. (2004)	28	0.6	8	6	5	47
Park et al. (2004)	21	NR	8	6	6	41
Rotman et al. (2004)	22	0.5	6	6	5	38
Wong et al. (2004)	27	0.7	6	6	3	43
Stevenson et al. (2004)	27	0.7	8	6	7	50
Stevenson et al. (2005)	28	0.8	10	7 <sup>d</sup>	5 <sup>d</sup>	51

Notes:

(a) Values given in the TAR are compared with those used in the more recent model studies of Table 7.4.5. None of these model studies includes the NO<sub>x</sub> source from oxidation of NH<sub>3</sub>, which could contribute up to 3 Tg-N yr<sup>-1</sup> according to the TAR. The source of NO<sub>x</sub> from STE is less than 1 Tg-N yr<sup>-1</sup> in all models, and this is well constrained from observations of N<sub>2</sub>O-NO<sub>y</sub> correlations in the lower stratosphere (Olsen et al., 2001).

(b) Values used in the TAR OxComp intercomparison of global tropospheric ozone models. Ranges from the emission inventories compiled by the TAR are given in parentheses.

(c) Values in parentheses are from a more recent GEOS-CHEM version (Martin et al., 2003b)

(d) Values recommended in the Stevenson et al. (2005) model intercomparison of 25 models; individual models used sources in the range 5.5-8.0 Tg-N yr<sup>-1</sup> for soils and 3.7-7.0 Tg-N yr<sup>-1</sup> for lightning.

The close agreement in NO<sub>x</sub> sources between models in Table 7.4.6 is somewhat artificial, reflecting the use of similar inventories and parameterizations rather than actual constraints on our understanding. The lightning source is particularly uncertain (Nesbitt et al., 2000; Tie et al., 2002), yet is of great importance because of the high production efficiency of ozone in the tropical upper troposphere. The range of the global lightning NO<sub>x</sub> source used in models (3-7 Tg-N yr<sup>-1</sup>) is designed to match tropical observations of ozone and nitrogen oxides (Martin et al., 2002), although large uncertainties in the model simulations of deep convection and the vertical distribution of lightning emissions detract from the strength of this constraint. Process-based models tend to predict higher lightning emissions (5-20 Tg-N yr<sup>-1</sup>; Price et al., 1997).

One significant development for constraining NO<sub>x</sub> sources since the TAR has been the GOME satellite observations of tropospheric NO<sub>2</sub> columns (Richter and Burrows, 2002). Leue et al. (2001) first showed how these data could be used to estimate the magnitude of NO<sub>x</sub> emissions. A detailed analysis by Martin et al. (2003b) found that the GOME NO<sub>2</sub> data yield a best estimate of 38 Tg N yr<sup>-1</sup> for the global surface source of NO<sub>x</sub> in 1996-1997 with a factor 1.6 uncertainty (Figure 7.4.4). This is consistent with the surface sources (excluding lightning and aircraft) used in the current generation of global models (34-42 Tg-N yr<sup>-1</sup>). Martin et al. (2003b) found that the biomass burning source of NO<sub>x</sub> had to be significantly lower than 6 Tg-N yr<sup>-1</sup>, which is already at the low end of the range used in current models. They also pointed out large regional discrepancies with current fossil fuel combustion inventories in the Middle East, South Africa, and some other regions as shown in Figure 7.4.4. Richter et al. (2005) used 1996-2004 trends observed by GOME to deduce a 50% increase in NO<sub>x</sub> emissions over industrial areas of China. Martin et al. (2003b) argued that GOME would be insensitive to lightning NO<sub>x</sub> due to preferential partitioning of upper tropospheric NO<sub>x</sub> to NO during daytime, but a careful analysis by Boersma et al. (2005) shows that the GOME data can constrain the global lightning NO<sub>x</sub> source for 1997 to be in the range 1.1-6.4 Tg N yr<sup>-1</sup>.

[INSERT FIGURE 7.4.4 HERE]

Other significant precursors for tropospheric ozone are CO and non-methane volatile organic compounds (NMVOCs), including in particular biogenic isoprene. Satellite measurements of CO from the MOPITT instrument launched in 1999 (Edwards et al., 2004) have provided important new constraints for CO emissions, pointing in particular to an underestimate of Asian sources in current inventories (Kasibhatla et

1 al., 2002; Petron et al., 2004; Arellano et al., 2004; Heald et al., 2004), as confirmed also by aircraft  
2 observations of Asian outflow (Palmer et al., 2003a; Allen et al., 2004). Recent measurements of biogenic  
3 NMVOC emissions and related ecosystem properties have been assimilated by Guenther et al. ( ) to construct  
4 updated global emission inventories. Satellite measurements of formaldehyde columns from the GOME  
5 instrument (Chance et al., 2000) have been used to place independent constraints on isoprene emissions and  
6 indicate values consistent in general with current inventories though with significant regional discrepancies  
7 (Palmer et al., 2003b; Shim et al., 2005).

8  
9 A few recent studies have explored the effect of aerosols on global tropospheric ozone involving both  
10 heterogeneous chemistry and perturbations to actinic fluxes. Jacob (2000) reviewed the heterogeneous  
11 chemistry involved. Hydrolysis of  $N_2O_5$  in aerosols is a well-known sink for  $NO_x$ , but other processes  
12 involving reactive uptake of  $HO_2$ ,  $NO_2$ , and  $O_3$  itself could also be significant. Martin et al. (2003b) found  
13 that including these processes along with actinic aerosol effects in a global CTM reduced ozone production  
14 rates by 6% globally, with larger effects over aerosol source regions (Tie et al., 2005).

15  
16 Overall, the current generation of tropospheric ozone models is generally successful in describing the  
17 principal features of the present-day global ozone distribution on the basis of underlying processes, although  
18 there are significant weaknesses (Brunner et al., 2003). Much less confidence is to be had in the ability of  
19 these models to reproduce the changes in ozone associated with perturbations to emissions or climate, the  
20 latter to be discussed below. The inability of current models to simulate observed long-term trends in ozone  
21 concentrations over the 20th century is of significant concern (Mickley et al., 2001; Hauglustaine and  
22 Brasseur, 2001; Shindell and Favulegi, 2002; Fusco and Logan, 2003; Shindell et al., 2003; Lamarque et al.,  
23 2005). Increasing our confidence in ozone models will require new approaches to model evaluation,  
24 involving for example correlations of ozone with other chemical or with meteorological variables.

#### 25 26 *7.4.4.2 Effects of climate change*

27 Climate change can affect tropospheric ozone in a number of ways through changes in precursor emissions,  
28 chemistry, and transport. A detailed report on ozone-climate interactions by the European Commission  
29 (2003) identifies changes in lightning, biomass burning, biogenic VOC emissions, humidity, and transport  
30 (including STE) as having potentially major impacts on ozone (Figure 7.4.5). These and other effects are  
31 discussed below. They could represent positive or negative feedbacks to climate change.

32  
33 [INSERT FIGURE 7.4.5 HERE]

#### 34 35 *7.4.4.2.1 Effects on ozone precursor emissions*

36 Climate change affects the sources of ozone precursors through physical response (lightning), biological  
37 response (soils, vegetation, biomass burning), and human response (energy generation, land use, agriculture).  
38 It is generally expected that lightning will increase in a warmer climate (Price and Rind, 1994a; Brasseur et  
39 al., 2005), though a GCM study by Stevenson et al. (2005) for the 2030 climate finds no global increase but  
40 instead a shift from the tropics to mid-latitudes. Perturbations to lightning could have a large effect on ozone  
41 in the upper troposphere (Toumi et al., 1996; Thompson et al., 2000; Martin et al., 2002; Wong et al., 2004).  
42 Mickley et al. (2001) found that observed long-term trends in ozone over the past century might be  
43 explainable by a concurrent increase in lightning.

44  
45 Biomass burning in the tropics and at high latitudes is likely to increase with climate change, both as a result  
46 of increased lightning and as a result of increasing temperatures and dryness (Price and Rind, 1994b; Stocks  
47 et al., 1998; Williams et al., 2001; Brown et al., 2004). Biomass burning is known to make a large  
48 contribution to the budget of ozone in the tropical troposphere (Thompson et al., 1996), and there is evidence  
49 that boreal forest fires can enhance ozone throughout the extratropical northern hemisphere (Jaffe et al.,  
50 2004). With climate warming, it is likely that boreal fires will increase due to a shorter duration for the  
51 seasonal snowpack and decreased soil moisture (Kasischke et al., 1995).

52  
53 The effect of climate change on biogenic VOC emissions is potentially large but complex. From an ozone  
54 perspective the effect on methane is most important and was discussed in Section 7.3.1. The effect on  
55 NMVOCs was examined by Constable et al. (1999) and Sanderson et al. (2003b). Although biogenic  
56 NMVOC emissions increase with increasing temperature, both studies concluded that ecosystem structural  
57 responses unfavorable to NMVOC emissions would compensate for the effect of warming,

#### 7.4.4.2.2 *Effects on chemistry*

Changes in temperature, humidity, and UV radiation intensity brought about by climate change could affect ozone significantly. GCM simulations by Stevenson et al. (2000) for the 21st century indicate a decrease in the lifetime of tropospheric ozone of 30% by 2100 as increasing water vapor enhances the ozone sink from the  $O(^1D) + H_2O$  reaction. They find that the ozone radiative forcing over the 1990–2100 period in the TAR SRES A2 scenario drops from 0.43 to 0.27  $W\ m^{-2}$  when the effect of climate change on ozone chemistry is included. Similar qualitative trends are found by Stevenson et al. (2005a) in an intercomparison of nine models for 2030 vs. 2000 climate.

#### 7.4.4.2.3 *Effects on transport*

Changes in atmospheric circulation could have a major effect on tropospheric ozone. Recent GCM studies concur that the STE source of tropospheric ozone should increase in the future climate because of the stronger Brewer-Dobson stratospheric circulation (Sudo et al., 2003; Collins et al., 2003; Zeng and Pyle, 2003; Stevenson et al., 2005b). Changes in vertical transport within the troposphere are also important, in view of the rapid increase in both ozone production efficiency and ozone lifetime with altitude. Convection is expected to intensify as climate warms (Rind et al., 2001), although this might not be the case in the tropics (Stevenson et al., 2005b). The implications are complex, as recently discussed by Pickering et al. (2001), Lawrence et al. (2003), Doherty et al. (2005), and Li et al. (2005). On the one hand, convection brings down ozone-rich air from the upper troposphere to the lower troposphere where it is rapidly destroyed, and replaces it with low-ozone air. On the other hand, injection of  $NO_x$  to the upper troposphere greatly increases its ozone production efficiency. Compensation between these two effects can either lead to a decrease or increase of ozone, depending on  $NO_x$  levels in the lower troposphere.

Changes in the modes of the general circulation can also affect the distribution and lifetime of tropospheric ozone. TOMS satellite observations of tropical tropospheric ozone columns show a strong interannual sensitivity to ENSO (Ziemke et al., 1999; Chandra et al., 2003), as illustrated in Figure 7.4.6, and this is also found in ozone models (Peters et al., 2001). Warm phase conditions lead to increased ozone over the equatorial western tropical Pacific, reflecting contributions from both Indonesian biomass burning and reduced upwelling (Sudo and Takahashi, 2001). In the central and eastern Pacific, by contrast, warm-phase conditions lead to depleted ozone because of weaker subsidence (Figure 7.4.6). The ENSO effect on tropospheric ozone is mainly confined to the tropics; at northern mid-latitudes, the Arctic Oscillation (AO) has been found recently to be a major contributor to tropospheric ozone variability (Creilson et al., 2003; Lamarque and Hess, 2004).

[INSERT FIGURE 7.4.6 HERE]

### 7.4.5. *The Hydroxyl Radical (OH)*

#### 7.4.5.1 *Present-day OH budget*

The hydroxyl radical (OH) is the primary cleansing agent of the lower atmosphere and it provides the dominant sink for many greenhouse gases (e.g.,  $CH_4$ , HCFCs, HFCs) and pollutants (e.g., CO, non-methane hydrocarbons). The steady-state lifetime of these trace gases is determined by the morphology of their atmospheric distribution, the kinetics of their reaction with OH, and the OH distribution. The local abundance of OH is mainly controlled by the local abundances of  $NO_x$ , CO,  $CH_4$  and higher hydrocarbons,  $O_3$ , water vapour as well as the intensity of solar ultraviolet radiation (UVR) at wavelengths shorter than 0.310  $\mu m$ . The primary source of tropospheric OH is a pair of reactions that start with the photodissociation of  $O_3$  by solar UVR:



Additionally, in the remote troposphere and in particular in the upper troposphere, photodissociation of oxygenated volatile organic chemicals such as peroxides, acetone and other ketones, alcohols, and aldehydes and may provide dominant sources of OH radical (e.g., Jaeglé et al., 2001; Tie et al., 2003; Singh et al., 2004). In urban environments or within the forest canopy, measurements also suggest that the processing of

1 unsaturated hydrocarbons, photolysis of carbonyls or HONO can also sustain a large pool of radicals (e.g.,  
2 Handisides et al., 2003; Heard et al., 2004). OH reacts with many atmospheric trace gases, in most cases as  
3 the first and rate-determining step of a reaction chain that leads to more or less complete oxidation of the  
4 compound. These chains often lead to formation of an HO<sub>2</sub> radical, which then reacts with O<sub>3</sub> or NO to  
5 recycle back to OH. Tropospheric OH and HO<sub>2</sub> are lost through radical-radical reactions leading to the  
6 formation of peroxides or with NO<sub>2</sub> to form HNO<sub>3</sub>. The sources and sinks of OH involve most of the fast  
7 photochemistry of the troposphere.

#### 9 7.4.5.2 Changes in OH over the past decades

##### 10 7.4.5.2.1 Top-down estimates

11 The global distribution of OH radicals cannot be observed directly because of the difficulty in measuring its  
12 small concentrations (about 10<sup>6</sup> OH molecules cm<sup>-3</sup> on average during daylight in the lower free troposphere)  
13 and because of strong variability of OH with geographical location, time of day, and season. However,  
14 indirect estimates of the average OH concentration can be obtained from observations of atmospheric  
15 concentrations of trace gases such as methyl chloroform (CH<sub>3</sub>CCl<sub>3</sub>) that are removed mostly by reaction with  
16 OH (with rate constants known from laboratory studies), and whose emission history is relatively well-  
17 known. The lifetime of CH<sub>3</sub>CCl<sub>3</sub> is then often used as a reference number to derive the lifetime of other  
18 species and, by convention, provides a measure of the global OH burden. Prinn et al. (2001) analyzed a 22-  
19 year record of global CH<sub>3</sub>CCl<sub>3</sub> measurements and emission estimates and inferred a large interannual and  
20 interdecadal variability in global OH. They estimated that OH values in 2000 were below those in the late  
21 1970s by 10 ± 24 %. Prinn et al. (2005) have extended in time their former analysis over the period 1979–  
22 2003. Their results suggest that global average OH levels have remained fairly steady from 1979 to 2003 with  
23 only a small maximum around 1989 and a larger minimum around 1998, with OH concentrations in 2003  
24 being comparable to those in 1979. Krol and Lelieveld (2003) calculated a variation of OH of +12% during  
25 the 1978–1990 period, followed by a decrease slightly larger than 12% in the decade 1991–2000. Over the  
26 entire 1978–2000 period that study provides an overall change close to zero. Bousquet et al. (2005) have also  
27 re-examined the OH trend over the 1980–2002 period from the inversion of CH<sub>3</sub>CCl<sub>3</sub> atmospheric  
28 observations accounting for varying meteorological fields over the considered period. They also infer a  
29 substantial OH inter-annual variability and a trend of –0.9% yr<sup>-1</sup> over the entire period in agreement with  
30 previous estimates (Figure 7.4.7). In a recent study, Manning et al. (2005) have used measured carbon  
31 monoxide containing radiocarbon (<sup>14</sup>CO) to derive the OH trend. They found no significant long-term trend  
32 in OH concentrations over 1989–2003 period but provide evidence for short-term variations of about 10%  
33 persisting for a few months. As a summary of these various analyses, it appears that the phase of the OH  
34 fluctuations (positive trend in the 1980s and a negative trend since the early 1990s) is more robustly  
35 determined by the inversions than their amplitude (*see* Section 2.3.3 for discussion).

36  
37 [INSERT FIGURE 7.4.7 HERE]

##### 39 7.4.5.2.2 Bottom-up analyses

40 The fluctuations in global OH derived from CH<sub>3</sub>CCl<sub>3</sub> or CO measurements are in conflict with observed  
41 methane growth rates and with forward model calculations. Karlsdottir and Isaksen (2000) used a 3D CTM  
42 accounting for varying nitrogen oxides (NO<sub>x</sub>), carbon monoxide (CO) and non-methane hydrocarbon  
43 (NMHC) emissions and found a positive trend in OH of +0.43% yr<sup>-1</sup> over the period 1980–1996. Dentener et  
44 al. (2003ab) used a 3D CTM accounting for varying emissions of ozone precursors and methane,  
45 meteorology and ozone column and derived a positive trend of +0.26% yr<sup>-1</sup> over the 1979–1993 period.  
46 Wang et al. (2004) have also used a 3D CTM accounting for interannual variations in methane emissions,  
47 transport, and column ozone to analyze the trend of methane from 1988 to 1997. They did not account for  
48 interannual variability of a number of other variables that affect OH such as concentrations of NO<sub>x</sub>,  
49 tropospheric ozone and NMHCs. They also derive a positive trend in OH over the considered period of  
50 +0.63% yr<sup>-1</sup>. Their calculated trend in OH is primarily associated with the negative trend in the overhead  
51 column ozone over the considered period and reduced to +0.16% yr<sup>-1</sup> when the total O<sub>3</sub> column is held  
52 constant.

#### 54 7.4.5.3 Longer-term changes in OH

##### 55 7.4.5.3.1 Impact of emissions

56 Because of its dependence on CH<sub>4</sub> and other pollutants, tropospheric OH is also expected to have changed  
57 since the pre-industrial era and to change in the future. Pre-industrial OH is likely to have been different than

today, but because of the counteracting effects of higher CO and CH<sub>4</sub> (decreasing OH) and increased NO<sub>x</sub> and O<sub>3</sub> (increasing OH) there is still little consensus on the magnitude of this change. Several model studies suggest a decline in weighted global mean OH from pre-industrial time to present-day of less than +10% (Shindell et al., 2001; Lelieveld et al., 2002a; Lamarque et al., 2005). Other studies have reported larger decreases in global OH of -16% (Mickley et al., 1999), -25% (Wong et al., 2004) and -33% (Hauglustaine and Brasseur, 2001). The model study by Lelieveld et al. suggests that during the past century OH concentration decreased substantially in the marine troposphere by reaction with CH<sub>4</sub> and CO, however, on a global scale it has been compensated by an increase over the continents associated with strong emissions of nitrogen oxides. As far as future changes in OH are concerned, IPCC (Prather et al., 2001), using scenarios reported in the IPCC Special Report on Emissions Scenarios (Nakicenovic et al., 2000) and on the basis of a comparison of results from 14 models, predicts that global OH could decrease by 10% to 18% by 2100 for 5 emission scenarios and increase by 5% for one scenario assuming large decreases in CH<sub>4</sub> and other ozone precursor emissions. Based on a different emission scenario, Wang and Prinn (1999) also predicted an OH decrease of 16 ± 3% in 2100.

#### 7.4.5.3.2 Effects of climate change

In addition to the emission changes future increases in direct and indirect greenhouse gases could also induce changes in OH through direct participation in OH-controlling chemistry, indirectly through stratospheric ozone changes that could increase solar ultraviolet in the troposphere, and potentially through changes in temperature, humidity, and clouds or climate change effects on biogenic emissions of methane and other ozone precursors. Changes in tropospheric water could have important chemical repercussions. The reaction between water vapour and electronically excited oxygen atoms constitutes the major source of tropospheric OH (Eq. 7.2). So, in a warmer, and potentially wetter climate, the abundance of OH is expected to increase. This effect was already proposed by Pinto and Khalil (1991) to explain the variation of OH during the cold dry Last Glacial Maximum (LGM). This effect was quantified by Martinerie et al. (1995) who calculated that the change in water vapour during the LGM was responsible for a 7% decrease in global average OH concentration. Brasseur et al. (1998) and Johnson et al. (1999) estimated that in a warmer climate corresponding to a doubling in CO<sub>2</sub> concentration, the global and annual mean OH concentration increases by +7% and +12.5% respectively. More recently, Hauglustaine et al. (2005) used a climate-chemistry 3D model and calculated a 16% reduction in global OH from present day to 2100 accounting for changes in surface emissions solely. The effect of climate change and mainly of increased water vapour in this model is to increase global OH by 13%. In that study, the competing effects of emissions and climate change maintain the future global average OH concentration close to its present-day value. The importance of the water vapour distribution on global OH as also been illustrated by Lamarque et al. (2005). These authors showed that under reduced aerosol emissions the warmer and moister climate directly increases global OH concentration significantly.

Changes in lightning NO<sub>x</sub> emissions in a warming climate have also the potential to significantly affect OH. Labrador et al. (2004) have calculated that global OH is very sensitive to the magnitude of the lightning NO<sub>x</sub> emissions and increases by 10% and 23% when the global lightning source is increased respectively by a factor of 2 and 4 from a 5 TgN yr<sup>-1</sup> best estimate. A similar sensitivity of global to the lightning source has been estimated by Wang et al. (2004) who calculated a 10.6% increase in OH for a doubling of the source (from 3 to 6 TgN yr<sup>-1</sup>). Regarding the large uncertainty on lightning emissions and the sensitivity of OH to the total amount of N emitted, an improved understanding of this source appears important for our ability to accurately simulate OH and its changes over time.

#### 7.4.5.4 Consequences on lifetimes

As described in more detail in Velders et al. (2005), for a given trace gas, each relevant sink process contributes to the additive first-order total loss frequency,  $l$ , which is variable in space and time such as oxidation by OH. A local lifetime  $\tau_{\text{local}}$  can be defined as the inverse of  $l$  evaluated at a point in space ( $x, y, z$ ) and time ( $t$ ):

$$\tau_{\text{local}} = 1 / l(x,y,z,t) \quad (7.3)$$

The global instantaneous atmospheric lifetime of the gas is obtained by integrating  $l$  over the considered atmospheric domain. The integral must be weighted by the distribution of the trace gas on which the sink

1 processes act. Consider a distribution of the trace gas  $C(x,y,z,t)$ , a global instantaneous lifetime derived from  
 2 the budget can be defined as:

$$3 \quad \tau_{\text{global}} = \int C \, dv / \int C l \, dv \quad (7.4)$$

4 where  $dv$  is an atmospheric volume element. This expression can be averaged over a year to determine the  
 5 global and annually averaged lifetime. The global atmospheric lifetime characterizes the time required to  
 6 turn over the global atmospheric burden. Because the total loss frequency  $l$  is the sum of the individual sink  
 7 process frequencies,  $\tau_{\text{global}}$  can also be expressed in terms of process lifetimes:

$$8 \quad 1/\tau_{\text{global}} = 1/\tau_{\text{tropospheric OH}} + 1/\tau_{\text{photolysis}} + 1/\tau_{\text{other processes}} \quad (7.5)$$

9 It is convenient to consider lifetime with respect to individual sink processes limited to specific regions, e.g.,  
 10 the oxidation by reaction with OH in the troposphere. However, the associated burden must always be global  
 11 and include all communicating reservoirs in order for (Eq. 7.4) to remain valid. In (Eq. 7.4), the numerator is  
 12 therefore integrated over the whole atmospheric domain and the denominator is integrated over the domain  
 13 in which the individual sink process is considered. In the case of  $\tau_{\text{tropospheric OH}}$ , the convention is that  
 14 integration is performed over the tropospheric domain. The use of different domains or different definitions  
 15 for the troposphere can lead to differences of 10% in the calculated value (Lawrence et al., 2001).

16 The lifetimes can be determined in global models by simulating the injection of a pulse of that gas and  
 17 watching the decay of this added amount. This decay can be represented by a sum of exponential functions,  
 18 each with its own decay time. These exponential functions are the chemical modes of the linearised  
 19 chemistry-transport equations of a global model (Prather, 1996; 2002). In the case of a  $\text{CH}_4$  addition, the  
 20 longest-lived mode has an  $e$ -fold time of 12 yr, very close to the steady-state perturbation lifetime of  $\text{CH}_4$   
 21 described in more details in Prather et al. (2001) and Velders et al. (2005). In the case of a CO, HCFCs, or  
 22 HCs addition, this same mode is also excited, but at a much reduced amplitude depending on the amount of  
 23 gas emitted (Prather, 1996; Daniel and Solomon, 1998). The pulse of added CO, HCFCs, or HCs, by causing  
 24 the concentration of OH to decrease and thus the lifetime of  $\text{CH}_4$  to increase temporarily, causes a build-up  
 25 of  $\text{CH}_4$  while the added burden of the gas persists. After the initial period defined by the photochemical  
 26 lifetime of the injected trace gas, this built-up  $\text{CH}_4$  then decays in the same manner as would a direct pulse of  
 27  $\text{CH}_4$ . Thus, changes in the emissions of short-lived gases can generate long-lived perturbations as shown in  
 28 global models (Wild et al., 2001; Derwent et al., 2001). Changes in tropospheric  $\text{O}_3$  accompany the  $\text{CH}_4$   
 29 decay on a 12-year time scale as an inherent component of this mode, a key example of chemical coupling in  
 30 the troposphere. Thus, any chemically reactive gas, whether a greenhouse gas or not, will produce some  
 31 level of indirect greenhouse effect through its impact on atmospheric chemistry.

32 Since OH is the primary oxidant in the atmosphere of many greenhouse gases including methane and  
 33 hydrogenated halogen species, their lifetime in the atmosphere and hence impact on the climate system will  
 34 directly be affected by changes in OH. Several studies have shown that variations on an annual basis in the  
 35 chemical removal of methane by OH exert an important impact in the variability of the methane growth rate  
 36 (Johnson et al., 2002; Warwick et al., 2002; Wang et al., 2004). These studies show that variations in  $\text{CH}_4$   
 37 oxidation by OH contribute to a significant fraction of the observed variations in the annual accumulation  
 38 rate of methane in the atmosphere. In particular, the observed slowdown in the growth rate of methane in the  
 39 atmosphere is attributed to a combination of slower growth of sources and increases in OH and the 1992–  
 40 1993 anomaly in  $\text{CH}_4$  growth rate can be explained by fluctuations in OH and wetland emissions after the  
 41 eruption of Mount Pinatubo (Wang et al., 2004). The methane variability simulated by Johnson et al. (2002)  
 42 accounting only from OH sink processes also indicates that the El Niño-Southern Oscillation cycle is the  
 43 largest component of that variability. These findings are consistent with the variability of global OH  
 44 reconstructed by Prinn et al. (2005) and Manning et al. (2005) and strongly affected by large-scale wildfires  
 45 as in 1997–1998, by El Niño events and the Mt Pinatubo eruption.

46 The importance of climate change on tropospheric chemistry and on the future evolution of methane has also  
 47 been investigated in several studies. In most cases the future methane lifetime increases when emissions  
 48 increase and climate change is ignored (Brasseur et al., 1998; Stevenson et al., 2000; Prather et al., 2001;  
 49 Hauglustaine and Brasseur, 2001; Hauglustaine et al., 2005). This feature reflects the fact that increased  
 50 levels of  $\text{CH}_4$  and CO depress OH reducing the  $\text{CH}_4$  sink. However, climate warming increases the  
 51

1 temperature-dependant CH<sub>4</sub> oxidation rate coefficient (Johnson et al., 1999), and increases in water vapour  
2 and NO<sub>x</sub> concentrations tend to increase OH. In most cases, these effects partly offset or even exceed the  
3 methane lifetime increase due to emissions. As a consequence, the future methane lifetime calculated by  
4 Brasseur et al. (1998), Stevenson et al. (2000) and Hauglustaine et al. (2005) remains relatively constant  
5 (within a few %) over the 21st century. In their transient simulation over the period 1990–2100, Johnson et  
6 al. (2001) found a dominant effect of climate change on OH in the free troposphere so that the global  
7 methane lifetime declines from around 9 years in 1990 to around 8.3 years after by 2025 and does not  
8 change significantly thereafter. The direct consequence is that the calculated methane increase in 2100 is  
9 reduced by 27% when climate change is considered. A similar conclusion about the relatively constant  
10 methane lifetime was reached by Stevenson et al. (2005) in their model intercomparison. As a result of future  
11 changes in emissions, the methane lifetime simulated by 25 state-of-the art chemistry transport models  
12 increased by 3% in 2030 from an ensemble mean of  $8.7 \pm 1.3$  years for the present-day. Under the 2030  
13 warmer climate scenario the lifetime was reduced by 5% so that the total effect of both emission and climate  
14 changes was to reduce the methane lifetime by only 2%.

#### 16 *7.4.6 Stratospheric Ozone and Climate*

18 During the last two decades a negative trend in globally averaged total ozone has been observed. It is  
19 generally accepted that an increase in chlorine and bromine loading is a major cause of this trend. In  
20 addition, atmospheric concentration of well-mixed greenhouse gases has also increased. This subsection  
21 presents a synthetic review of our understanding of the interactions between stratospheric ozone and climate.  
22 For more detailed discussions, see the reports published recently by WMO (2003), Chapters 3 and 4, by the  
23 IPCC (Special Report on Ozone and Climate, Chapter 1), and by the European Commission (2003).

25 [INSERT FIGURE 7.4.8 HERE]

##### 27 *7.4.6.1 Interactions*

28 Figure 7.4.8 illustrates schematically the processes determining ozone climate interactions in the troposphere  
29 and stratosphere. Stratospheric ozone is affected by climate change through changes in tropospheric  
30 temperature, in the water vapour content and possibly in the circulation and dynamics (e.g., Granier and  
31 Shine, 1999; Stevenson et al., 2000; Johnson et al., 2001). An increase in the concentration of greenhouse  
32 gases (GHG), especially CO<sub>2</sub>, cools the stratosphere and alters the ozone distribution (Rosenfield et al.,  
33 2002). In most of the stratosphere, a decrease in temperature reduces ozone depletion leading to higher  
34 ozone amounts and a positive correction to the GHG induced cooling. However, in the polar regions, there is  
35 a positive feedback process whereby cooling leads to enhanced ozone destruction via chlorine and bromine.

37 Ozone is a greenhouse gas and absorbs UV radiation in the stratosphere. The absorption of UV provides the  
38 heating responsible for the observed increase of temperature with height above the tropopause. Changes in  
39 stratospheric temperatures, whether induced by ozone change or GHG change, alter the Brewer-Dobson  
40 circulation. This controls the rate at which long-lived molecules, such as GHGs, CFCs, and halons are  
41 transported from the troposphere to various levels in the stratosphere. There is also statistical evidence that  
42 changes in the stratospheric circulation propagate downwards into the troposphere (e.g., Baldwin and  
43 Dunkerton, 1999).

45 Additionally, climate is affected by changes in stratospheric ozone. Stratospheric ozone radiates infrared  
46 radiation down to the troposphere. For a given percentage change in the vertical structure of ozone, the  
47 largest dependence of the radiative forcing (RF) is in the vicinity of the tropopause (Wang et al., 1980; Lacis  
48 et al., 1990). The climate sensitivity, (i.e. the surface temperature response per unit RF) on the vertical  
49 structure of the ozone perturbation is model dependent, but ozone perturbations in the lower stratosphere  
50 appear to be most efficient (e.g., Stuber et al. 2001; Joshi et al., 2003). Past ozone depletion has thus induced  
51 surface cooling, which has approximately balanced the greenhouse warming resulting from increasing  
52 abundances of ozone depleting substances (ODSs) and their substitutes. Taking into consideration the net  
53 change in ozone in the troposphere and stratosphere, the positive radiative forcing due to tropospheric ozone  
54 changes has dominated the negative stratospheric forcing over the period 1750–2000. Over the period 1970–  
55 2000, however, the two forcings were approximately of equal importance, but opposite in sign. Part of the  
56 long-term observed UV changes are driven by changes in stratospheric ozone.

#### 7.4.6.2 *Past ozone changes*

Past ozone losses have been largest in polar regions during spring. For example, the ozone hole over Antarctica has occurred every spring since the early 1980s and is a recurring phenomenon (Fioletov et al., 2002). In the year 2002, the ozone hole was of shorter duration than normal due to a unique stratospheric warming event. This is not an indication of recovery in ozone amounts, but rather the result of this dynamical disturbance (e.g., Newman et al., 2004). Ozone destruction there is driven by climatologically low temperatures combined with high chlorine and bromine amounts produced from the photochemical breakdown of primarily man-made CFCs and halons. Similar losses, but of much smaller magnitude, have occurred over the Arctic due to the same processes during cold winters. Warm winters have been relatively unaffected. Ozone losses in other regions have occurred due to increases in nitrogen oxides (from nitrous oxide emissions) and heterogeneous chemical processes on the surface of atmospheric aerosols, again combined with high chlorine and bromine amounts. Mid-latitudes have been influenced by transport of ozone from polar and tropical regions, as well as from local chemistry (aerosols and nitrogen oxides).

#### 7.4.6.3 *Stratospheric water vapour trend*

Stratospheric water vapour is controlled to a high degree by temperatures near the tropical tropopause. Long-term observed changes show that the tropical tropopause has cooled slightly during the past two decades. Most climate models do not simulate an increase in water vapour, neither for the past nor for the future. Long-term changes in lower stratospheric water vapour are currently not known, as e.g. decadal-scale changes of water vapour in the lower stratosphere for the time period 1992–2002 are very different between balloon and satellite datasets near Boulder (Oltmans et al., 2000; Rosenlof et al., 2001; Randel et al., 2004). A positive (negative) water vapour trend, if present, is expected to give rise to a positive (negative) trend in total ozone of comparable magnitude.

#### 7.4.6.4 *Future ozone changes*

Continued increases in greenhouse gases and a slow decrease in atmospheric halogen loading are expected to occur in the future decades. Both influence the abundance of stratospheric ozone. Over Antarctica, ozone should start to recover within the next 10 years, whereas in the Arctic, the recovery should be start within the next two decades (Figure 7.4.9).

[INSERT FIGURE 7.4.9 HERE]

In the Arctic, the climate feedback associated with GHG increase on the ozone recovery should be more prominent than in the Antarctic. The recovery to the levels last observed in 1980 is not expected to occur until about 2040. Thereafter there may be some further ozone losses due to increases in nitrous oxide emissions. When recovery does finally occur, the vertical distribution of ozone will be different from that which was observed in the 1980s due to the sensitivity of the chemical processes to the atmospheric temperature. One of the major scientific conclusions of recent model studies (Austin et al., 2003) is that we do not expect a future Arctic ozone hole comparable to the hole observed in Antarctica.

#### 7.4.6.5 *Uncertainties in current coupled Chemistry-Climate Models (CCMs)*

Coupled chemical climate models (CCMs) have been used to simulate past ozone changes and to provide prediction of the future evolution of ozone in response of human activities. Only a limited number of these models have assessed the sensitivity of stratospheric ozone to natural forcings such as large volcanic eruptions, solar variability (11-year cycle), and the quasi-biennial oscillation (QBO). These mechanisms have significant impacts on processes that determine the dynamics and chemistry of the atmosphere. A more detailed investigation of the role of natural variability in past ozone changes and other atmospheric changes is therefore important for assessments of ozone changes. The largest uncertainties in current Climate Chemistry Models (CCMs) stem from the performance of the underlying dynamical model components. Other components of CCMs (i.e., chemistry and transport) have not yet been tested to the same extent. These models are often characterized by temperature biases, which lead to substantial errors in the calculated degree of ozone depletion. The major uncertainties are associated with the representation of the resolved and unresolved portions of the dynamical wave spectrum and the excitation of waves at resolved and unresolved scales (including gravity waves).

### 7.5 **Aerosol Particles and the Climate System**

1 Anthropogenic aerosol particles such as sulphate and carbonaceous aerosols have substantially increased the  
2 global mean burden of aerosol particles from preindustrial times to the present-day. Aerosol particles affect  
3 the climate system via the following physical mechanisms: First, they scatter and can absorb solar radiation.  
4 Second, they can scatter, absorb and emit thermal radiation. Third, aerosol particles act as cloud  
5 condensation nuclei (CCN) and ice nuclei (IN), thereby affecting the hydrological cycle and radiation  
6 processes. The first two mechanisms are referred to as direct effects and are discussed in detail in Chapter 2.  
7 The last one is referred to as an indirect effect. It will be the subject of this chapter together with other  
8 atmospheric properties influenced by aerosols (e.g. semi-direct effect, suppression of convection). Even  
9 though the semi-direct effect is a consequence of the direct effect of absorbing aerosols, it changes cloud  
10 properties in response to these aerosols. This aspect will be discussed here while its radiative effects are  
11 subject of Chapter 2.

### 12 13 **7.5.1 What Major Climatic Factors Control Natural Aerosol Emissions and Burdens?**

14  
15 Aerosol sources are more complex than greenhouse gas sources, because aerosols often are formed within  
16 the atmosphere. Clouds are at the same time a source and a sink for aerosols. They are an important factor  
17 for determining the size, composition, lifetime and degree of mixing of aerosols. Also, aerosols left behind  
18 from an evaporating cloud are more effective CCN in subsequent cloud formation events. Thus, the  
19 anthropogenic burden hethero depends on the climate itself. In addition natural emissions, such as wind  
20 driven dust and sea salt or emissions from the marine and terrestrial biosphere, also depend on climate. This  
21 complexity is lost if simple empirical relationships between anthropogenic emissions and aerosol radiative  
22 effects are used in global climate models. Some examples of climatic influences on natural aerosol emissions  
23 and burdens are discussed below while anthropogenic aerosol emissions are subject of Chapter 2. The  
24 importance of natural aerosols and their dependence on the parameterizations used in different climate  
25 models is highlighted in Figure 7.5.1 (Kinne et al., 2005).

26  
27 [INSERT FIGURE 7.5.1 HERE]

#### 28 29 **7.5.1.1 Dust**

30 Estimates of global source strength of bulk dust aerosol range from ~1000 to 3000 Mt yr<sup>-1</sup> (Andreae, 1995;  
31 Duce, 1995; Textor et al., 2005) with high spatial and temporal variability. Zhang et al. (1997) estimated that  
32 ~800 Mt of Asian dust is injected into the atmosphere annually, about 30% of which is re-deposited onto the  
33 deserts and 20% is transported over regional scales, while the remaining ~50% is subject to long-range  
34 transport to the Pacific Ocean and beyond. Uncertainties for the estimates of global dust emissions are over  
35 more than a factor of two (Zender et al., 2004) due to problems in validating and modeling the global  
36 emissions. The representation of the high wind tail of the wind speed distribution alone—responsible for  
37 most of the dust flux—leads to differences in emissions in one model by more than 30% (Timmreck and  
38 Schulz 2004). Observations suggest that African dust may have varied by a factor of four during 1960–2000  
39 (Chiapello et al., 2005), possibly due to variability of rainfall in the Sahel zone. Single model simulations  
40 suggest a decrease of dust emissions by 20–60% for 2100 due to changes in vegetation cover in response to  
41 precipitation, temperature and CO<sub>2</sub> fertilization (Mahowald and Luo, 2003). Natural mineral dust is the most  
42 important aerosol component in terms of aerosol burden and radiative impact and the modulation of its  
43 source due to climate factors is thus likely to be of importance. The radiative effect of dust, suggested for  
44 example to intensify the African Easterly Waves, may be a feedback mechanism between climate and dust  
45 (Jones et al., 2005b).

46  
47 Dust may alter the climate, which in turn can feed back upon dust emission. For example, radiative forcing  
48 by dust from anthropogenic sources alters the atmospheric circulation, which feeds back upon dust emission  
49 from natural sources. Perlwitz et al. (2001) estimated that this feedback changes the global dust load by  
50 roughly 15%. In addition to natural dust production, human activities have created another potential source  
51 for dust through the process known as desertification, but the contribution of desertification through human  
52 activities to global dust emission is very uncertain with the estimation varying from 50% (Tegen et al.,  
53 1996), to less than 10% (Tegen et al., 2004), to insignificant values (Ginoux, 2001; Prospero et al., 2002). A  
54 43-year estimation of Asian dust emissions reveals that meteorology and climate have a greater influence on  
55 Asian dust emissions and associated Asian dust storm occurrences than desertification (Figure 7.5.2; Zhang  
56 et al., 2003). Estimates of future changes in dust emissions under several climate and land-use scenarios  
57 suggest dust emissions may increase or decrease. Either way the effects of climate change are more

1 important in controlling dust emissions than changes in land-use (Tegen et al., 2004). High Asian dust-  
2 induced heating of the land relative to the ocean would tend to reduce the thermal gradient between the land  
3 and ocean, thereby weakening the Asian winter monsoon circulation, with a feedback to reduce the dust  
4 emission from its sources (Zhang et al., 2002).

5  
6 [INSERT FIGURE 7.5.2 HERE]

7  
8 In addition, the deposition of aerosols has impacts on global ecosystems. Deposition of mineral dust in the  
9 ocean plays an important role in the biogeochemical cycle of the oceans. Also for terrestrial ecosystems the  
10 input of trace elements by dust deposition is of essential importance. For example, it has been proposed that  
11 the vegetation of the Amazon basin is highly dependent on Saharan dust deposition as it provides  
12 phosphorus, necessary for the maintenance of long-term productivity (see Section 7.3).

#### 13 14 7.5.1.2 *Natural organic carbon*

15 Biogenic organic material is both directly emitted into the atmosphere and produced by volatile organic  
16 compounds (VOC). Primary emissions have been thought to be a relatively minor source but some studies  
17 suggest that these emissions could be much higher (Jaenicke, 2005; Folberth et al., 2005). Kanakidou et al.  
18 (2005) estimate a global biogenic secondary organic aerosol production of  $\sim 30 \text{ Tg yr}^{-1}$  and recognize the  
19 potentially large, but highly uncertain, flux of primary biogenic particles. They suggest that primary  
20 emissions are of limited importance for climate since they consist mostly of the coarse aerosol fraction.  
21 Annual global biogenic VOC emission estimates range from about 0.5 to 1.2 Pg. However, there is a large  
22 range (<5% to >90%) of organic aerosol yield for individual compounds and atmospheric conditions  
23 resulting in estimates of global annual secondary organic aerosol production from biogenic VOC of about 32  
24 Tg that range from 2.5 to 44.5 Tg of organic matter per year (Tsigardis and Kanakidou, 2003). All biogenic  
25 VOC emissions are highly sensitive to changes in temperature, and some emissions respond to changes in  
26 solar radiation and precipitation (Guenther et al., 1995). In addition to the direct response to climatic  
27 changes, biogenic VOC emissions are also highly sensitive to climate-induced changes in plant species  
28 composition and biomass distributions. Several studies have examined the response of global biogenic VOC  
29 emissions to climate change (e.g., Turner et al., 1991; Adams et al., 2001; Sanderson et al., 2003b). These  
30 model studies predict that solar radiation and climate induced vegetation change can affect emissions, but  
31 they do not even agree on the sign of the change. The impact of precipitation on biogenic VOC emissions is  
32 even more uncertain and has not been included in global studies. The substantial increase in emissions with  
33 increasing temperature ( $\sim 10\%$  increase per degree C) predicted by these studies is based on algorithms  
34 described by Guenther et al. (1993) which describe the response of emissions to short term changes in  
35 temperature. There is evidence of physiological adaptations to higher temperatures that would lead to a  
36 greater temperature response for long-term temperature changes (Guenther et al., 1999). The response of  
37 biogenic secondary organic carbon production to a temperature change, however, could be considerably  
38 lower than the response of biogenic VOC emissions since aerosol yields can decrease with increasing  
39 temperature.

40  
41 New evidence reveals the ocean as a source of organic matter from biogenic origin (O'Dowd et al., 2004;  
42 Leck and Bigg, 2005). These findings are, however, still too recent to extrapolate any global organic flux  
43 from the ocean surface.

#### 44 45 7.5.1.3 *Sea salt*

46 Sea salt aerosol is a key aerosol constituent of the marine atmosphere. Sea salt aerosol particles are  
47 hygroscopic and function as cloud condensation nuclei, affecting the formation of clouds and development  
48 of rain; they serve as sinks for reactive gases and small particles, possibly suppressing new particle  
49 formation; and they scatter light, affecting the albedo and radiative balance of Earth. Sea salt aerosol  
50 production is affected by meteorological or environmental factors that modify surface properties of the  
51 ocean, wave breaking, or the turbulence level in the ocean surface waters; the formation, size distribution,  
52 entrainment, rise velocity, gas exchange characteristics, or bursting of bubbles; or the formation,  
53 entrainment, or behaviour of drops. The major meteorological and environmental factors that have been  
54 proposed in this regard are wind speed, atmospheric stability and wind friction velocity, sea and air  
55 temperatures, present and prior rain or snow, and the amount and nature of surface-active materials in the  
56 near-surface ocean waters (Lewis and Schwartz, 2005). The average annual global sea-salt flux from 12  
57 models is estimated to be  $16300 \pm 200 \text{ Tg}$  (Textor et al., 2005) of which 15% is emitted into the fine mode.

1 These submicrometer particles contribute significantly to the background aerosol mass and number  
2 concentrations in the marine atmosphere and substantially to the total optical depth of the atmosphere over  
3 the open ocean.

#### 4 7.5.1.4 *Dimethylsulphide (DMS)*

5 DMS produced by phytoplankton is the most abundant form in which the ocean releases gaseous sulphur.  
6 DMS sea-air fluxes may vary by orders of magnitude in space and time depending mainly on DMS sea  
7 surface concentration and on wind speed. Estimates of the global DMS flux vary widely depending mainly  
8 on the utilized DMS sea surface climatology, sea-air exchange parameterization, and wind speed data,  
9 ranging from 16 Tg S/yr up to 54 Tg S/yr (see Kettle and Andreae, 2000 for a review). Up to now none of  
10 the transient climate studies include a description of the DMS cycle in the ocean. The response of the DMS  
11 emission to climate change could therefore be only assessed through changes in the sea-air exchange rate,  
12 which varies with wind speed and temperature. Penner et al. (2001) showed a small increase in DMS  
13 emissions between 2000 and 2100 (from 26.0 Tg-S yr<sup>-1</sup> to 27.7 Tg-S yr<sup>-1</sup>) using a constant DMS sea surface  
14 concentration field together with a constant monthly climatological ice cover. As discussed in Section  
15 7.5.1.4., Gabric et al. (2004) predicted an increase of the globally integrated DMS flux perturbation of +14%  
16 for a tripling of the pre-industrial carbon dioxide concentration.

#### 17 7.5.1.5 *Climatic factors controlling aerosol burdens*

18 Knowledge of the various primary and secondary aerosol sources, combined with the size and composition  
19 dependent aerosol sinks, chemical transformations and transport processes in principle allow prediction of  
20 the chemical and physical nature of the atmospheric aerosol and their amounts in the atmosphere (burden).  
21 Some of these controlling factors, such as chemical reaction rates, depend on climate factors, especially  
22 temperature. As these are relevant to biogeochemical cycles, some of them will be highlighted here.

23 The burden of carbonaceous aerosols is influenced by the conversion of hydrophobic carbon to hydrophilic  
24 carbon. While this conversion depends on condensation, coagulation and oxidation, it can reasonably well be  
25 approximated as a first order reaction with a constant 24-h half-life in the present-day climate (Croft et al.,  
26 2005; Park et al., 2005). However, one key factor in controlling the burden of secondary organic aerosols in  
27 global climate models is the gas-particle partitioning (Kanakidou et al., 2004). We cannot simply assume  
28 that all semi-volatile products always condense in the same proportion, because that depends on the presence  
29 of primary organic and inorganic aerosols as well as on temperature and relative humidity.

30 Lohmann and Feichter (1997) showed that the sulphate burden increased by 50% when an aerosol scheme is  
31 coupled to a cloud scheme in a global climate model. A positive feedback loop is established in which more  
32 sulphate aerosols decrease the precipitation formation rate, which in turn increases the lifetime of sulphate  
33 aerosols and results in more long-range transport of sulphate to remote regions where wet removal is less  
34 efficient. If, however, a fraction of the anthropogenic black carbon (BC) is considered to act as contact ice  
35 nuclei, the precipitation formation via the ice phase is enhanced, removing aerosols from the atmosphere and  
36 thus terminating the positive feedback loop. This mechanism can reduce the increase in anthropogenic  
37 aerosol burden from pre-industrial to present-day times by 38% to 58% (Lohmann, 2002).

38 Stier et al. (2005) concluded that the lifetime of sulphate aerosols decreased 10% from pre-industrial to  
39 present-day conditions, the black carbon lifetime decreased 23% and mineral dust 2%, whereas the lifetime  
40 of particulate organic matter increased 11%. The alterations in aerosol lifetimes cannot unambiguously be  
41 attributed to microphysical processes, as lifetimes are also affected by geographical shifts of the dominant  
42 aerosol source regions. Moreover, the burdens of different aerosol species do not add linearly. Close to the  
43 anthropogenic source regions, deviations from additivity are found up to 30% and 15% for the accumulation  
44 mode number burden and aerosol optical thickness, respectively. These results challenge the appropriateness  
45 of bulk aerosol models for addressing climate change.

#### 46 7.5.2 *How Do Aerosols (Indirect Effects) Modify Clouds and Precipitation?*

47 Aerosol particles act as cloud condensation nuclei (CCN) and ice nuclei (IN). These indirect effects are the  
48 subject of this section together with aerosol induced changes in large-scale circulation and convection. They  
49 can be subdivided into processes contributing to the indirect aerosol effect as summarized in Table 7.5.1.  
50 The cloud albedo effect is the only true indirect aerosol forcing and therefore is subject of Chapter 2. The  
51

1 other effects involve feedbacks and will be discussed here. All processes that decrease the cloud droplet size  
 2 per given liquid water content simultaneously lead to an increase in cloud albedo (cloud albedo effect) and  
 3 reduce the precipitation formation. Thus, they immediately affect cloud dynamics and prolong cloud lifetime  
 4 (cloud lifetime effect), making it difficult to separate these different effects from observations. Heating due  
 5 to black carbon can cause cloud droplet evaporation (semi-direct), which could counteract some of the  
 6 aerosol-induced cooling described above. It also affects static stability as discussed in Section 7.5.4.1. If  
 7 some aerosol particles initiate freezing in a supercooled cloud, this cloud will rapidly glaciate due to the  
 8 difference in vapour pressure over ice and water (Bergeron-Findeisen process). Unlike cloud droplets, these  
 9 ice crystals grow in an environment of high supersaturation with respect to ice, quickly reaching  
 10 precipitation size, and with that, turning a non-precipitating into a precipitating cloud (glaciation effect and  
 11 Section 7.5.2.3.). The thermodynamic effect refers to aerosol-induced increases in the intensity of convective  
 12 clouds due to more latent heat of freezing that increases the amount of precipitation from polluted clouds  
 13 (Section 7.5.2.5.). The surface energy budget effect (Section 7.5.3.1.) describes the aerosol-induced decrease  
 14 in surface solar radiation with consequences for evaporation and precipitation.

15  
 16 **Table 7.51a.** Overview of the different aerosol indirect effects and their sign of the radiative forcing at the  
 17 top-of-the atmosphere (TOA)  
 18

Effect	Cloud Types Affected	Process	Sign of Change in TOA Radiation	Potential Magnitude	Certainty
Cloud albedo effect	All clouds	For the same cloud water or ice content more but smaller cloud particles reflect more solar radiation	Negative for water clouds, positive or negative for ice clouds	medium	certain
Cloud lifetime effect	All clouds	Smaller cloud particles decrease the precipitation efficiency thereby prolonging cloud lifetime	Negative	medium	indication
Semi-direct effect	All clouds	Absorption of solar radiation by soot leads to an evaporation of cloud particles and increases static stability	Positive or negative	small	potential
Glaciation indirect effect	Mixed-phase clouds	An increase in ice nuclei increases the precipitation efficiency	Positive	medium	potential
Thermodynamic effect	Mixed-phase clouds	Smaller cloud droplets inhibit freezing causing supercooled droplets to extend to colder temperatures	Positive or negative	medium	uncertain
Surface energy budget effect "Solar dimming"	All clouds	The aerosol induced increase in cloud optical thickness decreases the amount of solar radiation reaching the surface, changing the surface energy budget	n/a	n/a	indication

19  
 20 **Table 7.5.1b.** Overview of the different aerosol indirect effects and their implications for the shortwave  
 21 radiation at the surface  $F_{sfc}$  and for the hydrological cycle.  
 22

Effect	Sign of Change in $F_{sfc}$	Potential Magnitude	Certainty	Sign of Change in Precipitation	Potential Magnitude	Certainty
Cloud albedo effect	Negative	medium	certain	N/a	zero	certain
Cloud lifetime effect	Negative	medium	indication	Negative	small	indication
Semi-direct effect	Negative	large	potential	Negative	large	potential
Glaciation indirect effect	Positive	medium	potential	Positive	medium	potential
Thermodynamic effect	Positive or negative	medium	uncertain	Positive or negative	medium	uncertain
Solar dimming	Negative	large	indication	Negative	large	indication

### 7.5.2.1 *Observational aerosol effects on water clouds*

The indirect aerosol effect of changing cloud albedo is discussed in Chapter 2. Evidence of a cloud lifetime effect due to anthropogenic emissions of aerosols and their precursors stem, for instance, from the absence of a drizzle mode in ship tracks perturbing marine stratus cloud decks off the coast of California (Ferek et al., 1998) as well as from polluted versus clean clouds off the Atlantic coast of Canada (Peng et al., 2002). One remaining problem is that most climate models suggest an increase in liquid water when adding anthropogenic aerosols, whereas newer ship track studies show that polluted marine water clouds can have less liquid water than clean clouds (Platnick et al., 2000; Coakley et al., 2002). Ackerman et al. (2004) attribute this phenomenon to enhanced entrainment of dry air in polluted clouds in these instances.

On the individual cloud scale, the largest unknown is the contribution of organic aerosols as CCN (McFiggans et al., 2005 and Chapter 2). Recent studies that combine measurements with Lagrangian air parcel models off the coasts of Ireland, Eastern Canada and in the Arctic Ocean show that organics can increase the number of cloud droplets (O'Dowd et al., 2004; Lohmann and Leck, 2004; Leaitch et al., 2005). Over the Arctic Ocean this increase in cloud droplet number arises from the ability of the organic aerosols to lower the surface tension (Facchini et al., 1999; Decesari et al., 2003), whereas off the coasts of Ireland the cloud droplet increase is attributed to the addition of a new source of marine organics, mostly insoluble, active during phytoplankton blooms. Off the coast of Nova Scotia, the increase in cloud droplet number concentration is either caused by organics that are reasonably water-soluble and grow in a way similar to that of sulphate. Another possibility is that the organics are only weakly soluble and grow at a much slower rate than sulphate, which leads to a slightly higher cloud supersaturation and the activation of smaller aerosol particles. These chemical effects could be of the same order of magnitude as the cloud albedo effect (Nenes et al., 2002) and may be as important as unresolved cloud dynamics (Lance et al., 2004).

For the tropical Indian Ocean, Ramanathan et al. (2001) estimated a total indirect aerosol effect of  $-5 \text{ W m}^{-2}$  at the top-of-the-atmosphere (TOA) and of  $-6 \text{ W m}^{-2}$  at the surface. While the combined direct and semi-direct effect is negligible at the TOA, its surface forcing amounted to  $14 \text{ W m}^{-2}$ , pointing out the importance of considering surface radiative forcings in addition to TOA forcings.

### 7.5.2.2 *Global climate model estimates of the total anthropogenic aerosol effect*

The total anthropogenic aerosol effect as defined here includes estimates of the indirect cloud albedo and cloud lifetime effect. For those climate models that also simulate black carbon and link their aerosols to the radiation scheme, the total anthropogenic aerosol effect is the sum of the direct, semi-direct and indirect effects. The global mean direct and semi-direct effects at the TOA are, however, smaller than the indirect effects (Lohmann and Feichter, 2001; Penner et al., 2003). The influence of black carbon is dominated via its absorption of solar radiation within the atmosphere, which also leads to a large negative global mean forcing of  $-1.2$  to  $-4 \text{ W m}^{-2}$  at the surface (Ramanathan et al., 2001a; Lohmann and Feichter, 2001; Liepert et al., 2004).

Climate model estimates of the total anthropogenic aerosol effect at the top-of-the atmosphere are generally larger than estimated from inverse models (Anderson et al., 2003; Lohmann and Feichter, 2005). Climate model estimates of the importance of the cloud lifetime effect as compared with the cloud albedo effect are diverse. Whereas some models concluded that the cloud albedo effect is four times as important as the cloud lifetime effect other models simulate a cloud lifetime effect that is larger than the cloud albedo effect (Lohmann and Feichter, 2005). This discrepancy is independent of the chemical nature of the anthropogenic aerosol species that are used in these different simulations. Differences among the simulations include an empirical treatment between the aerosol mass and the cloud droplet number concentration versus a mechanistic relationship, the dependence of the indirect aerosol effect on the assumed background aerosol or cloud droplet number concentration and the competition between natural and anthropogenic aerosols as CCN (Ghan et al., 1998; O'Dowd et al., 1999). Likewise differences in the cloud microphysics scheme, especially in the autoconversion rate, cause uncertainties in estimates of the indirect aerosol effect (Lohmann and Feichter, 1997; Jones et al., 2001; Menon et al., 2002; 2003). Another problem in current climate models arises from neglecting particulate sulphate emissions. Adams and Seinfeld (2003) point out that particulate sulphate emissions are more efficient per unit mass than gas-phase emissions at increasing CCN concentrations. However, Stier et al. (2005) show that this effect is negligible when not just sulphate but the full suite of aerosol components is taken into account.

1  
2 The global mean total indirect aerosol effect, defined as the change in net radiation at TOA from pre-  
3 industrial times to present-day, is shown in Figure 7.5.3. The simulations by Lohmann (2004) and Lohmann  
4 and Diehl (2005) now include the influence of aerosols on the cloud droplet size distribution (dispersion  
5 effect as introduced by Liu and Daum (2002)) for the cloud albedo effect. The simulations by Rotstayn and  
6 Liu (2005) include the dispersion effect for the cloud albedo and the cloud lifetime effect (Liu et al., 2004).  
7 If the dispersion effect is taken into account, the total indirect aerosol effect is reduced by 15–35% (Peng and  
8 Lohmann, 2003; Rotstayn and Liu, 2003; 2005). This explains the small global mean total indirect aerosol  
9 effect in the simulation by Rotstayn and Liu (2005). The global mean indirect aerosol effect in the CAM-  
10 Oslo model has been reduced from  $-1.8 \text{ W m}^{-2}$  (Kristjansson, 2002) to  $-0.13 \text{ W m}^{-2}$  (Storelvmo et al., 2005),  
11 due to the introduction of microphysical sinks for cloud droplets and a cloud droplet activation scheme  
12 which accounts for the competition of the CCN for the available supersaturation.

13  
14 All models agree that the total indirect effect is larger over the Northern Hemisphere than over the Southern  
15 Hemisphere (Figure 7.5.3). The values of the total indirect aerosol effect on the Northern Hemisphere  
16 radiative forcing vary between  $-0.2$  and  $-4.4 \text{ W m}^{-2}$  and on the Southern Hemisphere between  $-0.1$  and  $-1.1$   
17  $\text{W m}^{-2}$ . Estimates of the ocean/land partitioning of the total indirect effect vary from 0.2 to 1.6. Values  
18 exceeding one are in better agreement with the estimate of the total indirect aerosol effect as derived from  
19 combining POLDER satellite data and the ECHAM4 GCM results (Lohmann and Lesins, 2002). However,  
20 the retrieval of clouds from the POLDER satellite is limited to clouds with a rather narrow cloud droplet size  
21 distribution (Rosenfeld and Feingold, 2003) and therefore may not be universally applicable. The agreement  
22 with the POLDER satellite data weakens when the cloud lifetime effect is ignored (Lohmann and Lesins,  
23 2002; Quaas et al., 2004), which provides indirect evidence for the existence of a cloud lifetime effect on a  
24 global scale. Similar conclusions were reached by Suzuki et al. (2004) when comparing simulations with and  
25 without a cloud lifetime effect with AVHRR satellite data of liquid water path as a function of column  
26 aerosol number (Nakajima et al. 2001).

27  
28 [INSERT FIGURE 7.5.3 HERE]

### 29 30 7.5.2.3 *Aerosol impacts on large-scale mixed-phase clouds*

31 Lohmann (2002) and Lohmann and Diehl (2005) show that if, in addition to mineral dust, hydrophilic black  
32 carbon aerosols are assumed to act as ice nuclei at temperatures between  $0^{\circ}\text{C}$  and  $-35^{\circ}\text{C}$ , then increases in  
33 aerosol concentration from pre-industrial times to present-day pose a glaciation indirect effect. Here  
34 increases in contact and immersion ice nuclei in the present-day climate result in more frequent glaciation of  
35 supercooled clouds and increase the amount of precipitation via the ice phase. This decreases the global  
36 mean cloud cover and leads to more absorption of solar radiation. The glaciation effect dominates if dust is  
37 assumed to be composed of kaolinite. On the contrary, if dust consists of montmorillonite, which is a better  
38 ice nucleus, then the climate impact by black carbon is smaller and the changes in the hydrological cycle are  
39 as expected from the warm indirect cloud lifetime effect (Lohmann and Diehl, 2005).

40  
41 Observations by Borys et al. (2003) in midlatitude orographic clouds show that for a given supercooled  
42 liquid water content, both the riming and the snowfall rates are smaller if the supercooled cloud has more  
43 cloud droplets. Examination of this effect in global climate model simulations with pre-industrial and  
44 present-day aerosol concentrations show that while the riming rate in stratiform clouds has indeed decreased  
45 due to the smaller cloud droplets in polluted clouds, the snowfall rate has actually increased. This is caused  
46 by the pollution induced increase in aerosol and cloud optical thickness, which reduces the solar radiation  
47 and causes a cooling that favours precipitation formation via the ice phase (Lohmann, 2004).

### 48 49 7.5.2.4 *Aerosol impacts on cirrus clouds*

50 A change in the number of ice crystals in cirrus clouds could also exert a cloud albedo effect in the same way  
51 that the cloud albedo effect acts for water clouds. In addition, a change in the cloud ice water content could  
52 exert a radiative effect in the infrared. The magnitude of these effects in the global mean has not yet been  
53 fully established, but the development of a physically based parameterization scheme of cirrus formation for  
54 use in global models led to significant progress in understanding underlying mechanisms of aerosol induced  
55 cloud modifications (Kärcher and Lohmann, 2002).

1 A global climate model study concludes that such an effect based solely on ubiquitous homogeneous  
2 freezing is small globally (Lohmann and Kärcher, 2002). This is expected to hold in the presence of  
3 heterogeneous ice nuclei (IN) that freeze at relative humidities over ice close to homogeneous values (above  
4 130–140%) (Kärcher and Lohmann, 2003). In situ measurements reveal that organic-containing aerosols are  
5 less abundant than sulphate aerosols in ice cloud particles, suggesting that organics do not freeze  
6 preferentially (Cziczo et al., 2004). A model study explains this finding by the disparate water uptake of  
7 organic aerosols, and suggests that organics are unlikely significantly to modify cirrus formation unless they  
8 are present in very high concentrations at low temperatures (compared with sulphate-rich particles) and  
9 hamper water condensation (Kärcher and Koop, 2004).

10  
11 With regard to aerosol effects on cirrus clouds, a strong link has been established between gravity wave  
12 induced, mesoscale variability in vertical velocities and climate forcing by cirrus (Kärcher and Ström, 2003).  
13 Hemispheric-scale studies of aerosol-cirrus interactions using ensemble trajectories suggest that changes in  
14 upper tropospheric cooling rates and ice-forming aerosols in a future climate may induce changes in cirrus  
15 occurrence and optical properties that are comparable in magnitude with observed decadal trends in global  
16 cirrus cover (Haag and Kärcher, 2004). Optically thin and subvisible cirrus are particularly susceptible to IN  
17 and therefore likely affected by anthropogenic activities.

18  
19 Radiative forcing estimates and observed trends of aviation-induced cloudiness are discussed in Section  
20 2.6.4. In terms of indirect effects on cirrus clouds developing from aircraft emissions, Lohmann and Kärcher  
21 (2002) show that the impact of aircraft sulphur emissions on cirrus properties via homogeneous freezing is  
22 small. Provided that black carbon particles from aviation serve as efficient IN, then maximum increases or  
23 decreases in ice crystal number concentrations of more than 40% are simulated in a climate model study  
24 assuming that the ‘background’ (no aviation impact) cirrus cloud formation is dominated by heterogeneous  
25 or homogeneous nucleation, respectively (Hendricks et al., 2005). Progress in assessing the impact of aircraft  
26 black carbon on cirrus is hampered by the poor knowledge of natural freezing modes in cirrus conditions and  
27 the inability to describe full complexity of cirrus processes in global models.

#### 28 29 *7.5.2.5 Aerosol impacts on convective clouds*

30 Rosenfeld (1999) and Rosenfeld and Woodley (2000) analyzed aircraft data together with satellite data  
31 suggesting that pollution aerosols suppress deep convective precipitation by decreasing cloud droplet size  
32 and delaying the onset of freezing. This hypothesis was confirmed with a cloud resolving model (Khain et  
33 al., 2001) such that supercooled cloud droplets down to  $-37.5^{\circ}\text{C}$  could only be simulated if the cloud  
34 droplets were small and numerous. On a global scale, Nuber et al. (2003) find large instantaneous local  
35 aerosol forcings that reduce the warm phase precipitation in convective clouds by this mechanism. The  
36 precipitation change at the surface is, however, guided by feedbacks within the system.

37  
38 Tropical biomass burning aerosols could lead to a reduction of ice crystal size in tropical deep convective  
39 clouds (Sherwood, 2002). These smaller and more numerous ice crystals would then lead to more scattering  
40 of solar radiation, i.e., exert a cloud albedo effect. They also evaporate more readily, thus increasing relative  
41 humidity. The increased relative humidity is partially transported into the stratosphere through the tropical  
42 pipe, which partially accounts for the increase of the stratospheric water vapour over the last 50 years  
43 (Rosenlof et al., 2001). Being a greenhouse gas, water vapour provides a positive radiative forcing that  
44 would partially offset the cloud albedo effect associated with the smaller ice crystal size in these deep  
45 convective clouds.

#### 46 47 *7.5.2.6 Aerosol impacts on precipitation and hydrological cycle*

48 CCN and IN are largely responsible for precipitation processes in clouds. Smoke from burning vegetation  
49 reduces cloud droplet sizes and delays the onset of precipitation (Warner and Twomey, 1967; Andreae et al.,  
50 2004; Rosenfeld, 1999) as do aerosols from urban and industrial air pollution (Rosenfeld, 2000). Also, desert  
51 dust suppresses precipitation in thin low altitude clouds (Mahowald and Kiehl, 2003; Rosenfeld et al., 2001).  
52 On the other hand, correlations between dust and ice clouds, as derived from satellite data, are not yet  
53 conclusive (Mahowald and Kiehl, 2003), although no concurrent surface precipitation measurements were  
54 provided in these studies. Only the recent study by Givarti and Rosenfeld (2004) indicates surface  
55 precipitation losses over topographical barriers by 15–25% downwind of major coastal urban areas in  
56 California and in Israel. On the contrary Jin et al. (2005) conclude from analyzing four years of satellite and  
57 in-situ observations that the change in urban rainfall amount in summer in New York and Houston is not

1 primarily caused by aerosols. Likewise Ayers (2005) concludes that rainfall anomalies over Australia cannot  
2 be linked to anthropogenic aerosols.

3  
4 Modelling studies largely suggest that anthropogenic aerosols suppress precipitation in the absence of giant  
5 CCN and aerosol-induced changes in ice microphysics (e.g., Lohmann, 2002; Menon and DelGenio, 2005)  
6 as well as in mixed-phase clouds where the ice phase only plays a minor role (Phillips et al., 2002). Giant sea  
7 salt nuclei, on the other hand, may override the precipitation suppression effect of the large number of small  
8 pollution nuclei (Feingold et al., 1999; Rosenfeld et al., 2002). Gong and Barrie (2003) predict a reduction of  
9 20–60% in the marine cloud droplet number concentrations (CDNC) (because of the presence of sea salt),  
10 with greatest reductions in the roaring 40s south (40–70%) and in the midlatitude north (20–40%). This  
11 reduction in CDNC enhances the precipitation formation. If these giant CCN were covered by film-forming  
12 compounds, then their impact would be less than previously estimated (Medina and Nenes, 2004).

13  
14 Khain et al. (2005) postulate that smaller cloud droplets, such as those originating from human activity,  
15 would reduce the production of drizzle drops. When these droplets freeze, the associated latent heat release  
16 results in more vigorous convection. In a clean cloud, on the other hand, drizzle would have left the cloud so  
17 that less latent heat is released when the cloud glaciates resulting in less vigorous convection. Therefore, no  
18 squall line is formed with maritime aerosol concentrations, but the squall line arises under the influence of  
19 higher continental aerosol concentrations and results in more precipitation after two hours of simulations  
20 with a detailed cloud microphysics model. More precipitation from polluted clouds is also simulated for  
21 different three-week periods over the Atmospheric Radiation Measurement Program (ARM) site in  
22 Oklahoma (Zhang et al., 2005) as well as for multicell cloud systems by Seifert and Beheng (2005). On the  
23 other hand, precipitation from single mixed-phase clouds is reduced under continental and maritime  
24 conditions when aerosol concentrations are increased (Khain et al., 2004; Seifert and Beheng, 2005).  
25 Modelling results of a thunderstorm in Florida suggest that the whole dynamic structure of the storms is  
26 influenced by varying dust concentrations (Van den Heever et al., 2004). In particular, the updrafts are  
27 consistently stronger and more numerous when Saharan dust is present compared with a clean air mass. This  
28 suggests that dust results in enhanced glaciation of convective clouds leading to dynamical invigoration of  
29 the clouds, larger amounts of processed water, and thereby enhanced rainfall at the ground (as discussed  
30 above). However, the precipitation enhancement lasted only 2 hours, after which precipitation decreased as  
31 compared with clean conditions. This highlights the complexity of the system and indicates that the sign of  
32 the global change in precipitation due to aerosols is not yet known.

33  
34 Global climate model estimates of the change in global mean precipitation due to all anthropogenic indirect  
35 aerosol effects are summarized in Figure 7.26. Consistent with the conflicting results from the detailed cloud  
36 system studies, the change in global mean precipitation varies between  $-0.03 \text{ mm day}^{-1}$  and  $+0.004 \text{ mm}$   
37  $\text{day}^{-1}$ . These differences are amplified over land, ranging from  $-0.12 \text{ mm day}^{-1}$  to  $0.02 \text{ mm day}^{-1}$ . Only  
38 Quaas et al. (2004) and Storelvmo et al. (2005) simulate an increase in land precipitation, which agrees best  
39 with the observed increase in terrestrial-mean precipitation trend in this century (Hulme et al., 1998).  
40 Continental precipitation did however decrease from 1947 to 1996, and especially from 1955 to 1970, before  
41 the clean-air acts were introduced in Europe and North America. This decrease in continental precipitation  
42 could be explained with indirect aerosol effects that are largest in the Northern Hemisphere (Figure 7.5.4).

43  
44 [INSERT FIGURE 7.5.4 HERE]

### 45 46 **7.5.3 How Are Changes in Aerosols/Clouds Affecting Processes at the Earth's Surface?**

#### 47 48 **7.5.3.1 Energy budget: decreased solar radiation**

49 By increasing aerosol and cloud optical depth, emissions of aerosols from human activity and their  
50 precursors cause a reduction of solar radiation at the surface (“solar dimming”). As such, worsening air  
51 quality contributes to regional aerosol effects. Such a reduction is observed many regions worldwide (remote  
52 sites like Mauna Loa and Arctic and Antarctic stations as well as industrial regions) (Gilgen et al., 1998;  
53 Liepert, 2002; Stanhill and Cohen, 2001; Wild et al., 2004). According to Liepert (2002) this decline of solar  
54 radiation from 1961 to 1990 amounts to 1.3% per decade mainly over land surfaces of the Northern  
55 Hemisphere, whereas the clear-sky atmospheric transmission in Chile is perturbed only after the El Chichon  
56 and Mt Pinatubo volcanic eruptions (Schwartz, 2005). This affects the partitioning of direct versus diffuse  
57 solar radiation: Liepert and Tegen (2002) concluded that over Germany both aerosol absorption and

1 scattering must have declined from 1975 to 1990 in order to explain the simultaneously weakened aerosol  
2 forcing and increased direct/diffuse solar radiation ratio. The direct/diffuse solar radiation ratio over the  
3 United States also increased from 1975 to 1990, here likely due to increases in absorbing aerosols.  
4 Increasing aerosol optical depth associated with scattering aerosols alone in otherwise clear skies produces a  
5 larger fraction of diffuse radiation at the surface, which results in larger carbon assimilation into vegetation  
6 (and therefore greater transpiration) without a substantial reduction in the total surface solar radiation  
7 (Niyogi et al., 2004).  
8

9 Over the Indian Ocean region during the dry winter monsoon season it has been estimated that  
10 anthropogenic aerosols, especially the highly absorbing aerosols, can decrease the average solar radiation  
11 absorbed by the surface in the range of 15 to 35 W m<sup>-2</sup> (Ramanathan et al., 2001b). This results in an  
12 increase in the atmospheric heating between the surface and 3 km altitude by as much as 60 to 100%. Similar  
13 perturbations in the atmosphere have been observed over other regions, namely East Asia, South America  
14 and sub-Saharan Africa, which are subjected to large loading of absorbing aerosols. Such a perturbation  
15 imposed over the Indian Ocean (in the 15°S–40°N and 50°E–120°E region) can lead to a large regional  
16 cooling at the surface in the range of 0.5 to 1°C accompanied by a warming of the lower troposphere by  
17 about 1°C, as shown in a GCM study with fixed ocean surface temperatures (Chung et al., 2002). This  
18 vertical heating gradient alters the latitudinal and inter-hemispheric gradients in solar heating; these gradients  
19 play a prominent role in driving the tropical circulation (Ramanathan et al., 2001b) and determining the  
20 amount of precipitation (Chung and Zhang, 2004).  
21

22 Global climate model estimates of the mean decrease in shortwave radiation at the surface in response to  
23 indirect aerosol effects vary between –1.6 and –3.1 W m<sup>-2</sup> (Figure 7.5.5). As for the TOA net radiation, the  
24 decrease is largest on the Northern Hemisphere with values approaching –5 W m<sup>-2</sup>. Consistent with the  
25 above-mentioned regional studies, most models predict larger decreases over land than over the oceans.  
26

27 [INSERT FIGURE 7.5.5 HERE]  
28

29 The decrease in solar radiation at the surface resulting from the increases in optical depth due to the direct  
30 and indirect anthropogenic aerosol effects is more important for controlling the surface energy budget than  
31 the greenhouse gas induced increase in surface temperature. This has been shown in equilibrium simulations  
32 with a global climate model coupled to a mixed-layer ocean model with increasing aerosol particles and  
33 greenhouse gases due to human activity from pre-industrial times to present-day (Liepert et al., 2004;  
34 Feichter et al., 2004), and in transient simulations (Roeckner et al., 1999). The conductive flux from below  
35 the surface is negligible in the long-term mean. The other components of the surface energy budget (thermal  
36 radiative flux, sensible and latent heat fluxes) decrease in response to the reduced input of solar radiation.  
37 This mechanism could explain the observations of decreased pan evaporation over the last 50 years reported  
38 by Roderick and Farquhar (2002). As global mean evaporation must equal precipitation in equilibrium  
39 climate simulations, a reduction in the latent heat flux leads to a reduction in precipitation (Liepert et al.,  
40 2004).  
41

42 Recent surface observations show that the long term decline in solar radiation at land surfaces turned into an  
43 increase in surface solar radiation during the 1990s (Wild et al., 2005), in agreement with recent emission  
44 reductions in the “old” industrial regions in the northern hemisphere (Krüger and Graßl, 2002) as well as  
45 with long-term black carbon trends in the Canadian Arctic (Sharma et al., 2004) and sulphate deposition  
46 declines over Europe and North America since 1978. This also led to an improvement in air quality in these  
47 areas. Although Pinker et al. (2005) detect an overall global brightening signal of 0.16 W m<sup>-2</sup> yr<sup>-1</sup> from  
48 satellite data between 1983 to 2001, that change is a combination of a decrease in surface solar radiation  
49 until about 1990, followed by a sustained increase. Thus, the increasing greenhouse effect may no longer be  
50 masked by an aerosol-induced decline in solar radiation, resulting in the enhanced warming observed since  
51 the 1990s. The decrease in global mean precipitation from pre-industrial times to the present may reverse  
52 into an increase in global mean precipitation of about 1% in 2021–2050 as compared with 1961–1990,  
53 because the increased warming due to black carbon and greenhouse gases will dominate over the sulphate  
54 cooling (Roeckner et al., 2005). In South Asia, absorbing aerosols in atmospheric brown clouds may have  
55 played a major role in the observed South Asian climate and hydrological cycle changes and may have  
56 masked as much as 50% of the surface warming due to the global increase in greenhouse gases (Ramanathan

1 et al. 2005). Their simulations raise the possibility that, if current trends in emissions continue, the South  
2 Asian subcontinent may experience a doubling of the drought frequency in future decades.

### 3 4 7.5.3.2 *Aerosol effects on the diurnal temperature range*

5 Hansen et al. (1995) found that, although the aerosol direct effect or an increase in continental cloud albedo  
6 could contribute to a dampening of the surface temperature diurnal range (DTR), only an increase in  
7 continental cloud cover would be consistent with observations (Karl et al., 1993). The required cloud  
8 increase depends on cloud height and would need to be of the order of 1% global coverage for low clouds  
9 (i.e., 2–5% over land). More recently, Piers and Solomon (2003) observed a “weekend effect” in DTR whose  
10 most likely cause is aerosol-cloud interactions. Over the industrialized part of China in winter the cloud  
11 lifetime effect increases the nighttime temperature due to enhanced downward longwave forcing, thus  
12 reducing DTR by 0.7°C (Huang et al., 2005).

### 13 14 7.5.3.3 *Interactions of aerosols with land surfaces*

15 As discussed in detail in Chapter 2, through absorption of solar radiation, deposition of black carbon on  
16 snow surfaces decreases the albedo of the snow, causing a further warming that will lead to more melting of  
17 snow. The global mean radiative impact of an increase in the snow and ice albedo of  
18 1.5% in the Arctic and 3% in Northern Hemisphere land areas yields an increase in climate forcing of 0.3 W  
19 m<sup>-2</sup> (Hansen and Nazarenko, 2004).

## 20 21 7.5.4 *How Are Aerosols Affecting Circulation Patterns?*

### 22 23 7.5.4.1 *Effects on stability*

24 Changes in the atmospheric lapse rate modify the longwave emission and affect the water vapour feedback  
25 (Hu, 1996) and the formation of clouds. Observations and model studies show that an increase in the lapse  
26 rate produces an amplification of the water vapour feedback (Sinha, 1995). As aerosols cool the Earth’s  
27 surface and warm the aerosol layer the lapse rate will decrease and suppress the water vapour feedback. Thus  
28 a more stable boundary layer suppresses greenhouse gas warming and may enhance aerosol cooling. On a  
29 global scale, Feichter et al. (2004) find that aerosol cooling in the free troposphere generally decreases the  
30 lapse rate if only anthropogenic aerosols are considered. However, the atmosphere becomes more stable if  
31 aerosols and greenhouse gases increase due to an aerosol cooling near the surface, especially in polluted  
32 regions of the Northern Hemisphere, and to greenhouse gas warming aloft. The change in atmospheric  
33 stability strongly depends on the altitude of the black carbon heating (Penner et al., 2003).

34  
35 Dust modelling results suggest the existence of a contrast in radiative forcing between continents and oceans.  
36 Depending on the assumptions and the dust loading used in the calculation, the continental heating change  
37 could range from –4 to +10 W m<sup>-2</sup> (Wang et al., 2004) to 0 to +20 W m<sup>-2</sup> (Weaver et al., 2002) to +20 to +40  
38 W m<sup>-2</sup> (Sokolik and Toon, 1996) while the oceanic cooling could range from 0 to –4 W m<sup>-2</sup> (Wang et al.,  
39 2004) to 0 to –18 W m<sup>-2</sup> (Weaver et al., 2002) to –35 W m<sup>-2</sup> to –15 W m<sup>-2</sup> (Sokolik and Toon, 1996). The  
40 dust-induced thermal contrast changes between the Eurasian continent and the surrounding oceans are also  
41 found either to trigger or modulate a rapidly varying or unstable Asian winter monsoon circulation (Zhang et  
42 al., 2002). Heating of a lofted dust layer could also increase the occurrence of deep convection (Stephens et  
43 al., 2004).

44  
45 Aerosols also cause a contrast of radiative forcing between TOA and surface, as deduced from SeaWiFS  
46 satellite data. Mean clear-sky solar radiative heating for the winters of 1998 and 1999 has also been found to  
47 decrease at the ocean surface by 12 to 30 W m<sup>-2</sup>, but only by 4 to 10 W m<sup>-2</sup> at the top of the atmosphere over  
48 the tropical northern Indian Ocean. This threefold difference (due largely to solar absorption by soot) and the  
49 large magnitude of the observed negative surface forcing both imply that tropical aerosols might increase the  
50 strength of the thermal inversion and slow down the hydrological cycle (Satheesh and Ramanathan, 2000;  
51 Ramanathan et al., 2005). This contrast between TOA and surface during the period of the large Indonesian  
52 fires (September–December 1997) may also have enhanced the atmospheric stability, weakened the  
53 atmospheric circulation, and augmented drought conditions (Chou et al., 2002).

### 54 55 7.5.4.2 *Effects on the large-scale circulation*

56 Through the aerosol radiation-circulation feedback, the scattering and absorption of radiation by aerosols can  
57 cause atmospheric pressure and circulation changes, which have the potential, for example, to modify Arctic

1 teleconnection patterns like the Barents Sea Oscillation (Rinke et al., 2004). The decadal to interannual  
2 variability of sea-salt aerosol concentrations in northern Greenland ice cores also identifies the eastern and  
3 northeastern Pacific region as the most important centre of action responsible for variations in sea-salt  
4 aerosol export onto the Greenland ice sheet which is related to the Pacific/North American teleconnection  
5 pattern (Fischer 2001). The influence of aerosols and other anthropogenic pollutants, through land-sea  
6 interaction (differential heating), topography (Western Ghats), and horizontal thermal gradients (SST versus  
7 land surface temperature) on the coastal circulations over Equatorial Indian Ocean, can produce significant  
8 diurnal variability and heterogeneity that may have interactive feedback teleconnections between local and  
9 large scale features (Mohanty et al., 2001).

10  
11 Large tropical volcanic eruptions produce winter warming patterns over Northern Hemisphere continents  
12 forced by gradients of radiative heating from sulphate aerosols in the lower stratosphere. These effects must  
13 be considered for accurate dynamical seasonal predictions of Northern Hemisphere winter temperature over  
14 both North America and Eurasia (Robock, 2001).

15  
16 Menon et al. (2002b) find that circulation changes due to different types of aerosols can vary. In a model  
17 simulation with absorbing aerosols, the upper-level westerlies to the north and easterlies to the south of the  
18 Tibetan plateau are stronger. The simulation with scattering aerosols yields weaker anticyclones with  
19 westerlies present south of the plateau. These results indicate that absorbing aerosols, mainly black carbon,  
20 can reduce the solar radiation reaching the surface and can warm the atmosphere, affecting the vertical  
21 temperature profile, latent heat fluxes, atmospheric stability and convection. These changes in convection  
22 can in turn modify the large-scale atmospheric circulation as shown in Figure 7.5.6. In India and China,  
23 where aerosols have been added, increasing rising motions are seen while comparable increased subsidence  
24 to the south and north are present in simulations. Wang (2004) models the effect of black carbon on climate  
25 and finds that BC heats the atmosphere mainly between 5°N and 50°N, corresponding to the most polluted  
26 areas. The radiative effect due to BC in the model alters the meridional sensible and latent heat transport,  
27 convective fluxes especially over the tropics, and hence the atmospheric circulation. In the northern part of  
28 the intertropical convergence zone (ITCZ) deep convection is enhanced due to BC while south of ITCZ the  
29 strength of the convection is substantially reduced. The precipitation change is found to depend mainly on  
30 the imposed atmospheric solar heating. Figure 7.5.7 shows the percentage change in precipitation revealing  
31 more clearly the regions of enhanced and suppressed precipitation (Chung et al., 2002) over the Indian  
32 Ocean region: the effects spread beyond the region of imposed forcing. Over arid regions in Southwest Asia  
33 the precipitation decreases. The atmospheric lifetime of aerosols increases due to suppression of  
34 precipitation. The drier conditions resulting from suppressed rainfall can induce more dust and smoke due to  
35 the burning of drier vegetation (Ramanathan et al., 2001a), thus affecting both regional and global  
36 hydrological cycles (Wang, 2004).

37  
38 [INSERT FIGURE 7.5.6 HERE]

39  
40 [INSERT FIGURE 7.5.7 HERE]

41  
42 In Southern China, increases in both greenhouse gases and anthropogenic aerosols may be responsible for  
43 the observed droughts in recent years (Cheng et al., 2005), caused by a strengthening of the West Pacific  
44 Subtropical High in the early summer over the last 40 years, with the high-pressure system extending further  
45 westward over the continent in Southern China. Because the early summer average temperature contrast  
46 between the land and ocean decreases, the southwesterly monsoon from the ocean onto mainland China  
47 weakens and a surface horizontal wind divergence anomaly occurs over Southern China stabilizing the  
48 boundary layer. Thus, less moisture is transported to Southern China, causing the drying trend.

49  
50 Observed precipitation trends over land for the period 1900–1998 show a complex pattern in the tropics  
51 indicating, for example, a drying of the Sahel in North Africa (Hulme et al., 1998). Dry conditions in the  
52 Sahel are associated with a near-global, quasi-hemispheric pattern of contrasting sea surface temperature  
53 anomalies (cooler in the northern hemisphere and warmer in the southern hemisphere). Using a global  
54 climate model/mixed-layer ocean model, Williams et al. (2001) and Rotstayn and Lohmann (2002) show that  
55 the dynamical and hydrological changes in this region in response to the indirect effect of anthropogenic  
56 sulphate aerosols are similar to the observed changes that have been associated with the Sahelian drought  
57 (Giannini et al., 2003). In addition, aerosol induced cooling of the Mediterranean Sea surface has reduced

1 evaporation, which has decreased moisture transport to North Africa and the Middle East (Lelieveld et al.,  
2 2002b). That is, the anthropogenic aerosol cooling reduces the solar radiation and the surface temperature  
3 predominately in the Northern Hemisphere, causing a southward shift of the ITCZ and a weakening of the  
4 African summer monsoon (Figure 7.5.8). If, on the contrary, the Northern Hemisphere surface temperature  
5 increases more than the Southern Hemisphere surface temperature due to the increase in fossil fuel  
6 combustion of black carbon, then the ITCZ shifts northward, strengthens the Indian summer monsoon and  
7 increases the rainfall in the Sahel (Roberts and Jones, 2004).

8  
9 [INSERT FIGURE 7.5.8 HERE]

10  
11 In summary, an increase in aerosol load decreases air quality and the amount of solar radiation reaching the  
12 surface. This negative forcing competes with the greenhouse gas warming for determining the change in  
13 evaporation (Schwartz, 1993; Roeckner et al., 1999 and Feichter et al., 2004). While model simulations  
14 suggest that global mean evaporation and precipitation have decreased from pre-industrial to present-day  
15 times when considering both greenhouse gases and aerosols (Liepert et al., 2004), in future both evaporation  
16 and precipitation could increase (Roeckner et al., 2005).

## 17 18 **7.6 Concluding Remarks**

19  
20 Biogeochemical cycles interact closely with the climate system over a variety of temporal and spatial scales.  
21 An illustration of this interaction on geological timescales is provided by the Vostok ice core record, which  
22 provides a dramatic evidence of the coupling between the carbon cycle and the climate system. The  
23 dynamics of the Earth system that can be inferred from this record results from a combination of external  
24 forcing (in this case long-term periodic changes in the orbital parameters of the earth and hence of the energy  
25 intercepted by our planet) and a large array of feedback mechanisms within the earth environment (see  
26 Chapter 6). On smaller timescales, a wide range of forcings that originate from human activities (conversion  
27 and fragmentation of natural ecosystems, emissions of greenhouse gases, nitrogen fixation, degradation of  
28 air quality, stratospheric ozone depletion) is expected to produce planetary-wide effects and perturb  
29 numerous feedback mechanisms that characterize the dynamics of the earth system.

30  
31 [START OF BOX 7.2]

### 32 33 **Box 7.2: Effects of Climate Change on Air Quality**

34  
35 Weather is a key variable affecting air quality. Surface air concentrations of pollutants are highly sensitive to  
36 boundary layer ventilation, winds, temperature, humidity, and precipitation. Anomalous hot and stagnant  
37 conditions in the summer of 1988 were responsible for the highest ozone year on record in the northeastern  
38 United States (Lin et al., 2001). The summer heat wave in Europe in 2003 was associated with exceptionally  
39 high ozone (Ordonez et al., 2005; Box 7.2, Figure 1). Such high interannual variability of surface ozone  
40 correlated with temperature demonstrates the potential air quality implications of climate change over the  
41 next century.

42  
43 [INSERT BOX 7.2, FIGURE 1 HERE]

44  
45 Only a few GCM studies have investigated how air pollution meteorology might respond to future climate  
46 change. Rind et al. (2001) found that increased continental ventilation as a result of more vigorous  
47 convection should decrease surface concentrations, while Holzer and Boer (2003) found that weaker winds  
48 should result in slower dilution of pollution plumes and hence higher concentrations. A more focused study  
49 by Mickley et al. (2004) for the eastern United States found an increase in the severity and persistence of  
50 regional pollution episodes due to the reduced frequency of ventilation by cyclones tracking across Canada.  
51 This effect more than offset the dilution associated with the small rise in mixing depths. A decrease in  
52 cyclone frequency at northern mid-latitudes has indeed been noted in observations from the past few decades  
53 (McCabe et al., 2001).

54  
55 A number of studies in the United States have shown that summer daytime ozone concentrations correlate  
56 strongly with temperature (NRC, 1989). This correlation appears to reflect contributions of comparable  
57 magnitude from (1) temperature-dependent biogenic VOC emissions, (2) thermal decomposition of

1 peroxyacetylnitrate (PAN) which acts as a reservoir for NO<sub>x</sub>, and (3) association of high temperatures with  
2 regional stagnation (Jacob et al., 1993; Sillman and Samson, 1995). Empirical relationships between ozone  
3 air quality standard exceedances and temperature, as shown in Box 7.2, Figure 2, integrate all of these effects  
4 and could be used to estimate how projected regional changes in temperature would affect ozone air quality.  
5 However, the ozone background may respond differently, decreasing with increasing temperatures because  
6 of associated increases in humidity (Stevenson et al., 2005b). Changes in solar irradiance at the surface due  
7 to changes in cloudiness and recovery of the ozone layer could also have a significant impact on ozone  
8 production in polluted regions.

9  
10 [INSERT BOX 7.2, FIGURE 2 HERE]

11  
12 A few GCM studies have examined more specifically the effect of changing climate on ozone air quality,  
13 assuming constant emissions. Knowlton et al. (2004) used a GCM coupled to a regional climate model  
14 (RCM) to investigate the impact of 2050 vs. 1990 climate change on ozone concentrations in the New York  
15 City metropolitan area. They found a significant ozone increase that they translated into a 4.5% increase in  
16 ozone-related acute mortality. Langner et al. (2005) used a RCM driven by two different GCMs to examine  
17 changes in the AOT40 statistic (ozone-hours above 40 ppbv) over Europe in 2050–2070 relative to present.  
18 They found an increase in southern and central Europe, and a decrease in northern Europe, that they  
19 attributed to different regional trends in cloudiness and precipitation. Dentener et al. (2005) found in an  
20 intercomparison of nine GCM ozone simulations for 2030 vs. 2000 climate that the annual mean surface  
21 ozone concentrations over populated continents decreased slightly, by ~1 ppbv. Trends in annual mean  
22 concentrations may be driven more by decreases in the global ozone background (Stevenson et al., 2005b)  
23 than by increases the frequency and severity of high-ozone episodes.

24  
25 There has been less work on the sensitivity of aerosols to meteorological conditions. Regional model  
26 simulations by Aw and Kleeman (2003) find that increasing temperatures should increase surface aerosol  
27 concentrations due to increased production of aerosol precursors (in particular semi-volatile organic  
28 compounds and nitric acid) although this is partly compensated by the increasing vapor pressure of these  
29 compounds at higher temperatures. Perturbations to precipitation frequencies and patterns might be expected  
30 to have a major impact on aerosol concentrations, but the GCM study by Mickley et al. (2004) for 2000–  
31 2050 climate change finds in fact relatively little effect in the United States.

32  
33 Intercontinental transport of pollution is increasingly recognized as an important issue for regional air  
34 quality. Climate change would be expected to affect this transport through perturbation to the general  
35 circulation. A model study by Li et al. (2002) found that transatlantic transport of pollution from North  
36 America to Europe is correlated with the positive phase of the North Atlantic Oscillation (NAO), and such a  
37 correlation is apparent in TOMS observations of tropospheric ozone columns over the northeast Atlantic  
38 (Creilson et al., 2003). Perturbation to the NAO phase resulting from climate change would thus be expected  
39 to have a major effect on transatlantic transport of pollution.

40  
41 [END OF BOX 7.2]

42  
43 The interactions between physical, chemical, and biological processes and feedback mechanisms that  
44 provide the conditions necessary for life on earth, are not yet fully understood or quantified, but will have to  
45 be more accurately represented in the future generations of climate models. The response of the climate  
46 system to anthropogenic forcing is expected to be more complex than a simple cause-effect relationship;  
47 rather, it could exhibit chaotic behavior with cascades of effects across the different scales and with the  
48 potential for abrupt and perhaps irreversible transitions.

49  
50 [INSERT FIGURE 7.6.1 HERE]

51  
52 This chapter has assessed how processes related to vegetation dynamics, carbon exchanges, gas-phase  
53 chemistry and aerosol microphysics could affect the climate system. These processes, however, cannot be  
54 considered in isolation because of the potential interactions that exist between them. A striking example  
55 highlighting the need to consider the links between the carbon cycle and aerosols is provided by Andreae et  
56 al. (2005). These authors suggest that future climate change in response to atmospheric CO<sub>2</sub> increase  
57 depends strongly on the intensity of present-day aerosol cooling. They stress that our insufficient

1 understanding of the present climate forcing of aerosols limits our ability to quantify the carbon-climate  
2 feedback and hence to perform accurate climate projections for the future. Figure 7.6.1 shows that future  
3 climate warming projected by a model in response to anthropogenic CO<sub>2</sub> emissions should be considerably  
4 more pronounced when the present-day aerosol cooling is assumed to be large than when it is ignored.  
5 Brasseur and Roeckner (2005) estimate that the hypothetical removal from the atmosphere of the entire  
6 burden of anthropogenic sulphate aerosol particles would produce a rather immediate increase of about  
7 0.8°C in the globally averaged temperature with geographical patterns that bear resemblance with the  
8 temperature changes found in greenhouse gas scenario experiments (Figure 7.6.2). Thus, environmental  
9 strategies aimed at maintaining ‘global warming’ below a prescribed threshold must therefore account not  
10 only for CO<sub>2</sub> emissions but also for measures implemented to improve air quality.

11  
12 [INSERT FIGURE 7.6.2 HERE]

13  
14 In order to cope with the complexity of earth system processes and their interactions, and particularly to  
15 evaluate sophisticated models of the earth system, observations and long-term monitoring of climate and  
16 biogeochemical quantities will be essential. Climate models will have to reproduce accurately important  
17 processes and feedback mechanisms that are discussed in the present chapter.  
18  
19

1 **References**

- 2
- 3 Achard, F., H. Eva, H.J. Stibig, P. Mayaux, J. Gallego, and T. Richards, 2002: Determination of  
4 deforestation rates of the world's humid tropical forests. *Science*, 297, 999-1002.
- 5 Ackerman, A.S., O.B. Toon, D.E. Stevens, A.J. Heymsfield, V. Ramanathan, and E.J. Welton, 2000:  
6 Reduction of tropical cloudiness by soot. *Science*, 288, 1042-1047.
- 7 Ackerman, A.S., M.P. Kirkpatrick, D.E. Stevens, and O.B. Toon, 2004: The impact of humidity above  
8 stratiform clouds on indirect climate forcing. *Nature*, 432, 1014-1017.
- 9 Adams, J., J. Constable, A. Guenther, and P. Zimmerman, 2001: An estimate of natural volatile organic  
10 compound emissions from vegetation since the last glacial maximum, *Chemosphere- Global Change*  
11 *Science*, 3, 73-91.
- 12 Adams, P. J., and J. H. Seinfeld, 2003: Disproportionate impact of particulate emissions on global  
13 condensation nuclei concentrations. *Geophysical Research Letters*, 30,1239,  
14 doi:10.1029/2002GL016303.
- 15 Adler, R.F., G.J. Huffman, A. Chang, R. Ferraro, P.-P. Xie, J. Janowiak, B. Rudolf, U. Schneider, S. Curtis,  
16 D. Bolvin, A. Gruber, J. Susskind, P. Arkin, and E. Nelkin, 2003: The version-2 Global Precipitation  
17 Climatology Project (GPCP) monthly precipitation analysis (1979-present). *Journal of*  
18 *Hydrometeorology*, 4, 1147-1167.
- 19 Allredge, A.L., U. Passow, and B.E. Logan, 1993: The abundance and significance of a class of large,  
20 transparent organic particles in the ocean. *Deep-Sea Research I*, 40(6), 1131-1140.
- 21 Allen, D., K. Pickering, and M. Fox-Rabinovitz, 2004: Evaluation of pollutant outflow and CO sources  
22 during TRACE-P using modelcalculated, aircraft-based, and Measurements of Pollution in the  
23 Troposphere (MOPITT)-derived CO concentrations. *Journal of Geophysical Research*, 109, D15S03,  
24 doi:10.1029/2003JD004250.
- 25 Anderson, T.L., R.J. Charlson, S.E. Schwartz, R. Knutti, O. Boucher, H. Rodhe, and J. Heintzenberg, 2003:  
26 Climate forcing by Aerosols - a hazy picture. *Science*, 300, 1103-1104.
- 27 Andreae, M. O., 1995: Climate effects of changing atmospheric aerosol levels. In: *World Survey of*  
28 *Climatology, Future Climates of the World* [Henderson-Sellers, A. (ed.)]. Amsterdam, Elsevier, pp 341-  
29 392.
- 30 Andreae, M.O., and P. Merlet, 2001: Emission of trace gases and aerosols from biomass burning. *Global*  
31 *Biogeochemical Cycles*, 15, 955– 966.
- 32 Andreae, M.O., D. Rosenfeld, P. Artaxo, A.A. Costa, G.P. Frank, K.M. Longo, and M.A.F. Silvas-Dias,  
33 2004: Smoking rain clouds over the Amazon. *Science*, 303, 1337-1342.
- 34 Andreae, M.O., C.D. Jones, and P.M. Cox, 2005: Strong present-day aerosol cooling implies a hot future.  
35 *Nature*, 435(7046), 1187-1190.
- 36 Angert, A., S. Biraud, C. Bonfils, C.C. Henning, W. Buermann, J. Pinzon, C.J. Tucker, and I. Fung, 2005:  
37 Drier summers cancel out the CO<sub>2</sub> uptake enhancement induced by warmer springs. *Proceedings of the*  
38 *National Academy of Sciences USA*. 102, 10823-10827.
- 39 Angert, A., S. Biraud, C. Bonfils, W. Buermann, and I. Fung, 2004: CO<sub>2</sub> seasonality indicates origins of  
40 post-Pinatubo sink. *Geophysical Research Letters*, 31(11), L11103, doi:10.1029/2004GL019760.
- 41 Archer, D., 2005: The fate of fossil fuel CO<sub>2</sub> in geologic time. *Journal of Geophysical Research*, (in press).
- 42 Archer, D., H. Kheshgi, and E. Maier-Reimer, 1998: Dynamics of fossil fuel CO<sub>2</sub> neutralization by marine  
43 CaCO<sub>3</sub>. *Global Biogeochemical Cycles*, 12(2), 259-276.
- 44 Archer, D.A., A. Winguth, D. Lea, and N. Mahowald, 2000: What caused the glacial/interglacial  
45 atmospheric pCO<sub>2</sub> cycles? *Reviews of Geophysics*, 38, 159-189.
- 46 Arellano, A.F., Jr., P.S. Kasibhatla, L. Giglio, G.R. van der Werf, and J.T. Randerson, 2004: Top-down  
47 estimates of global CO sources using MOPITT measurements. *Geophysical Research Letters*, 31,  
48 L01104, doi:10.1029/ 2003GL018609.
- 49 Armstrong, R. A., C. Lee, J. I. Hedges, S. Honjo, and S. G. Wakeham, 2002: A new, mechanistic model for  
50 organic carbon fluxes in the ocean based on the quantitative association of POC with ballast minerals.  
51 *Deep-Sea Research II*, 49, 219-236.
- 52 Arora, V., 2005: Comment on “Optimized stomatal conductance of vegetated land surfaces and its effects on  
53 simulated productivity and climate” by A. Kleidon. *Geophysical Research Letters*, 108, L08708,  
54 doi:10.1029/2004GLO22110.
- 55 Arora, V.K. and G. J. Boer, 2003: A representation of variable root distribution in dynamic vegetation  
56 models. *Earth Interactions*, 7(6), 1-19.

- 1 Arora, V.K. and G.J. Boer, 2005a: A parameterization of leaf phenology for the terrestrial ecosystem  
2 component of climate models, *Global Change Biology*, 11(1), 39-59.
- 3 Arora, V.K. and G.J. Boer, 2005b: Simulating competition and coexistence between plant functional types in  
4 a dynamic vegetation model. *Earth Interactions Journal*, (submitted).
- 5 Artaxo, P., Maenhaut, W., Storms, H. e Van Grieken, R., 1990: Aerosol characteristics and sources for the  
6 Amazon Basin during the wet season. *Journal of Geophysical Research*, 95, 16971-16985.
- 7 Aulakh, M.S., R. Wassmann, and H. Rennenberg, 2001: Methane emissions from rice fields—quantification,  
8 mechanisms, role of management, and mitigation options. *Advances in Agronomy*, 70, 193-260.
- 9 Austin, J., D. Shindell, S.R. Beagley, C. Brühl, M. Dameris, E. Manzini, T. Nagashima, P. Newman, S.  
10 Pawson, G. Pitari, E. Rozanov, C. Schnadt, and T.G. Shepherd, 2003: Uncertainties and assessments of  
11 chemistry-climate models of the stratosphere. *Atmospheric Chemistry & Physics*, 3, 1-27.
- 12 Avissar, R., and D. Werth, 2005: Global hydroclimatological teleconnections resulting from tropical  
13 deforestation. *Journal of Hydrometeorology*, 6(2), 134–145.
- 14 Avissar, R., P.L. Silva Dias, M.A.F. Silva Dias, C. Nobre, 2002: The Large-scale Biosphere-Atmosphere  
15 Experiment in Amazonia (LBA): Insights and future research needs. *Journal of Geophysical Research*,  
16 107(D20), 8034, doi:10.1029/2002JD002507.
- 17 Aw, J., and M.J. Kleeman, 2003: Evaluating the first-order effect of intraannual air pollution on urban air  
18 pollution. *Journal of Geophysical Research*, 108, 4365, doi:10.1029/2002JD002688.
- 19 Ayers, G.P., 2005: "Air pollution and climate change: has air pollution suppressed rainfall over Australia?"  
20 *Clean Air and Environmental Quality*, 39, 51-57.
- 21 Bacastow, R.B., 1976: Modulation of atmospheric carbon dioxide by the Southern Oscillation. *Nature*, 261,  
22 116–118.
- 23 Bacastow, R.B., and C.D. Keeling, 1981: Atmospheric carbon dioxide concentration and the observed  
24 airborne fraction. In: *Carbon Cycle Modelling* [Bolin, B. (ed.)]. SCOPE 16. John Wiley and Sons, New  
25 York.
- 26 Bacastow, R.B., J.A. Adams, C.D. Keeling, D.J. Moss, T.P Whorf, C.S. Wong, 1980: Atmospheric carbon-  
27 dioxide, the southern oscillation, and the weak 1975 el-Niño. *Science*, 210(4465), 66-68.
- 28 Baida Roy, S., C.P. Weaver, D.S. Nolan, and R. Avissar, 2003: A preferred scale of landscape forced  
29 mesoscale circulations? *Journal of Geophysical Research*, 108(D22), 8854,  
30 doi:10.1029/2002JD003097.
- 31 Baker, T.R., O.L. Phillips, Y. Mahli, S. Almeida, L. Arroyo, A. Di Fiori, N. Higuchi, T.J. Killeen, S.G.  
32 Laurance, W.F. Laurance, S.L. Lewis, A. Monteagudo, D.A. Neill, N.C. A. Pitman, N. Silva, and R.  
33 Vasquez Martinez, 2004: Increasing biomass in Amazonian forest plots. *Philosophical Transactions of  
34 the Royal Society of London, Series B*, 359, 353-365.
- 35 Bakker, D.C.E., Y. Bozec, P.D. Nightingale, L. Goldson, M.-J. Messias, H.J.W. de Barr, M. Liddicoat, I.  
36 Skjelvan, V. Strass, and A.J. Watson, 2005: Iron and mixing affect biological carbon uptake in SOIREE  
37 and EisenEx, two Southern Ocean iron fertilisation experiments. *Deep-Sea Research I*, 52, 1001-1019.
- 38 Balsam, W.L., 1983: Carbonate dissolution on the Muir Seamount (Western North Atlantic):  
39 interglacial/glacial changes. *Journal of Sedimentary Petrology*, 53, 719-731.
- 40 Barford, C.C., S.C. Wofsy, M.L. Goulden, J.W. Munger, E.H. Pyle, S.P. Urbanski, L. Hutyrá, S.R. Saleska,  
41 D. Fitzjarrald, and K. Moore, 2001: Factors controlling long and short term sequestration of  
42 atmospheric CO<sub>2</sub> in a mid-latitude forest. *Science*, 294(5547), 1688-1691.
- 43 Barlage, M., and X. Zeng, 2004: Impact of observed vegetation root distribution on seasonal global  
44 simulations of land surface processes. *Journal of Geophysical Research*, 109, D09101,  
45 doi:10.1029/2003JD003847.
- 46 Battle, M., M.L. Bender, P.P. Tans, J.W.C. White, J.T. Ellis, T. Conway, R.J. Francey, 2000: Global carbon  
47 sinks and their variability inferred from atmospheric O<sub>2</sub> and delta C-13. *Science*, 287(5462), 2467-2470.
- 48 Bauer, T.R., P.M. Williams, and E.R.M. Druffel, 1992: <sup>14</sup>C activity of dissolved organic carbon fractions in  
49 the north-central Pacific and Sargasso Sea. *Nature*, 357, 667-670.
- 50 Beauchamp, B., 2004: Natural gas hydrates: myths, facts, and issues. *Comptes Rendus Geoscience*, 336, 751-  
51 765.
- 52 Behrenfeld, M.J., and P.G. Falkowski, 1997: A consumer's guide to phytoplankton productivity models.  
53 *Limnology and Oceanography*, 42(7), 1479-1491.
- 54 Bell, G.D., M.S. Halpert, C.F. Ropelewski, V.E. Kousky, A.V. Douglas, R.C. Schnell, and M.E. Gelman,  
55 1999: Climate assessment for 1998. *Bulletin of the American Meteorological Society*, 85, S1-S48.
- 56 Benemann, J.R., 1992: The use of iron and other trace-element fertilizers in mitigating global warming.  
57 *Journal of Plant Nutrition*, 15(19), 2277-2313.

- 1 Berger, A., 2001: The role of CO<sub>2</sub>, sea-level and vegetation during the Milankovitch-forced glacial-  
2 interglacial cycles. In: *Proceedings Geosphere-Biosphere Interactions and Climate* [Bengtsson, L.  
3 (ed.)]. Pontifical Academy of Sciences, 9–13 November 1998, Vatican City.
- 4 Bertrand, C., and A. Royer, 2004: Aerosol optical depth spatio-temporal characterization over the Canadian  
5 BOREAS domain. *International Journal of Remote Sensing*, 25(15), 2903-2917.
- 6 Betts, A., 2004: Understanding hydrometeorology using global models. *Bulletin of the American*  
7 *Meteorological Society*, 85, 1673-1688.
- 8 Betts, A., J. Ball, and J. McCaughey, 2001: Near-surface climate in the boreal forest. *Journal of Geophysical*  
9 *Research*, 106, 33529-33541.
- 10 Betts, R., P. Cox, M. Collins, P. Harris, C. Huntingford, C. Jones, 2004: The role of ecosystem-atmosphere  
11 interactions in simulated Amazonian precipitation decrease and forest dieback under global change  
12 warming. *Theoretical and Applied Climatology*, 78(1-3), 157-175.
- 13 Bey, I., D.J. Jacob, R.M. Yantosca, J.A. Logan, B.D. Field, A.M. Fiore, Q. Li, H. Liu, L.J. Mickley, and  
14 M.G. Schultz, 2001: Global modeling of tropospheric chemistry with assimilated meteorology: model  
15 description and evaluation. *Journal of Geophysical Research*, 106(D19), 23073-23096.
- 16 Bishop, J.K.B., T.J. Wood, R.E. Davies, and J.T. Sherman, 2004: Robotic observations of enhanced carbon  
17 biomass and export at 55 degrees S during SOFeX. *Science*, 304(5669), 417-420.
- 18 Boersma, K.F., H.J. Eskes, E.W. Meijer, and H.M. Keider, 2005: Estimates of lightning NO<sub>x</sub> production  
19 from GOME satellite observations. *Atmospheric Chemistry & Physics Discussions*, 5, 3047-3104.
- 20 Bogner J.E., R.L. Sass, and B.P. Water, 2000: Model comparisons of methane oxidation across a  
21 management gradient: wetlands, rice production systems, and landfill. *Global Biogeochemical Cycles*,  
22 14, 1021-1033.
- 23 Bolin, B., and E. Eriksson, 1959: Changes in the carbon dioxide content of the atmosphere and sea due to  
24 fossil fuel combustion. In: *The atmosphere and sea in motion* [Bolin, B. (ed.)], Rossby memorial  
25 volume. Rockefeller Inst., New York, 130-142.
- 26 Bonan, G.B., 2001: Observational evidence for reduction of daily maximum temperature by croplands in the  
27 midwest United States. *Journal of Climate*, 14, 2430-2442.
- 28 Bonan, G.B., K.W. Oleson, M. Vertenstein, S. Levis, X. Zeng, Y. Dai, R.E. Dickinson and Z-L. Yang, 2002:  
29 The land surface climatology of the Community Land Model coupled to the NCAR Community  
30 Climate Model. *Journal of Climate*, 15, 3123-3149.
- 31 Bonan, G.B., S. Levis, L. Kergoat, and K.W. Oleson, 2002: Landscapes as patches of plant functional types:  
32 an integrating concept for climate and ecosystem models. *Global Biogeochemical Cycles*, 16,  
33 doi:10.1029/2000GB001360.
- 34 Bonan, G.B., S. Levis, S. Sitch, M. Vertenstein, and K.W. Oleson, 2003: A dynamic global vegetation model  
35 for use with climate models: concepts and description of simulated vegetation dynamics. *Global*  
36 *Change Biology*, 9, 1543-1566.
- 37 Bond, T.C., D.G. Streets, K.F. Yarber, S.M. Nelson, J.-H. Woo, and Z. Klimont, 2004: A technology-based  
38 global inventory of black and organic carbon emissions from combustion. *Journal of Geophysical*  
39 *Research*, 109, doi:10.1029/2003JD003697.
- 40 Bonfils, C., A. Angert, C.C. Henning, S. Biraud, S.C. Doney, and I. Fung, 2005: Extending the record of  
41 photosynthetic activity in the eastern United States into the presatellite period using surface diurnal  
42 temperature range. *Geophysical Research Letters*, 32(8), L08405, doi: 10.1029/2005GL022583.
- 43 Bonfils, C., I. Fung, S. Doney, and J. John, 2004: On the detection of summertime terrestrial photosynthetic  
44 variability from its atmospheric signature. *Geophysical Research Letters*, 31(9), L09207, doi:  
45 10.1029/2004GL019453.
- 46 Bopp, L., O. Aumont, P. Cadule, S. Alvain, and M. Gehlen, 2005: Response of diatoms distribution to global  
47 warming and potential implications – a global model study. *Geophysical Research Letters*, (submitted).
- 48 Bopp, L., P. Monfray, O. Aumont, J.L. Dufresne, H. Le Treut, G. Madec, L. Terray, and J.C. Orr, 2001:  
49 Potential impact of climate change on marine export production. *Global Biogeochemical Cycles*, 15(1),  
50 81-99.
- 51 Borken, W., E.A. Davidson, K. Savage, J. Gaudinski, and S.E. Trumbore, 2003: Drying and wetting effects  
52 on carbon dioxide release from organic horizons. *Soil Science Society of America Journal*, 67(6), 1888-  
53 1896.
- 54 Børshheim, K.Y., and S.M. Myklesstad, 1997: Dynamics of DOC in the Norwegian Sea inferred from monthly  
55 profiles collected during 3 years at 66 N, 2 E. *Deep-Sea Research I*, 44, 593-601.

- 1 Borys, R.D., D.H. Lowenthal, S.A. Cohn, and W.O.J. Brown, 2003: Mountaintop and radar measurements of  
2 anthropogenic aerosol effects on snow growth and snowfall rate. *Geophysical Research Letters*, 30,  
3 doi:10.1029/2002GL016855.
- 4 Bousquet, P., P. Peylin, P. Ciais, C. Le Quéré, P. Friedlingstein, and P.P. Tans, 2000: Regional changes in  
5 carbon dioxide fluxes of land and oceans since 1980. *Science*, 290(5495), 1342–1346.
- 6 Bousquet, P., D.A. Hauglustaine, P. Peylin, C. Carouge, and P. Ciais, 2005: Two decades of OH variability  
7 as inferred by an inversion of atmospheric transport and chemistry of methyl chloroform. *Atmospheric  
8 Chemistry & Physics*, (submitted).
- 9 Bouwman A. F., L. J. M. Boumans, and N. H. Batjes, 2002a: Estimation of global NH<sub>3</sub> volatilization loss  
10 from synthetic fertilizers and animal manure applied to arable lands and grasslands. *Global  
11 Biogeochemical Cycles*, 16(2), doi:10.1029/2000GB001389.
- 12 Bouwman A. F., L. J. M. Boumans, and N. H. Batjes, 2002b: Emissions of N<sub>2</sub>O and NO from fertilized  
13 fields: Summary of available measurement data. *Global Biogeochemical Cycles*, 16(4), 1058,  
14 doi:10.1029/2001GB001811.
- 15 Bouwman A. F., L. J. M. Boumans, and N. H. Batjes, 2002c: Modeling global annual N<sub>2</sub>O and NO  
16 emissions from fertilized fields, *Global Biogeochemical Cycles*, 16(4), 1080,  
17 doi:10.1029/2001GB001812.
- 18 Boyd, P.W., and S.C. Doney, 2002: Modelling regional responses by marine pelagic ecosystems to global  
19 climate change. *Geophysical Research Letters*, 29(16), doi:10.1029/2001GL014130.
- 20 Boyle, E.D., 1988: The role of vertical chemical fractionation in controlling late quaternary atmospheric  
21 carbon dioxide. *Journal of Geophysical Research*, 93(C12), 15701-15714.
- 22 Brasseur, G. and E. Roeckner, Impact of improved air quality on the future evolution of climate, *Geophys.  
23 Res. Lett.*, submitted 2005.
- 24 Brasseur, G.P., J.T. Kiehl, J-F. Müller, T. Schneider, C. Granier, X-X. Tie, and D. Hauglustaine, 1998: Past  
25 and future changes in global tropospheric ozone: impact on radiative forcing. *Geophysical Research  
26 Letters*, 25(20), 3807-3810.
- 27 Brasseur, G., M. Schultz, C. Granier, M. Saunio, T. Diehl, M. Botzet, E. Roeckner, and S. Walters, 2005:  
28 Impact of climate change on the future chemical composition of the global troposphere. *Journal of  
29 Climate*, (accepted).
- 30 Bréon, F.-M., D. Tanré, and S. Generoso, 2002: Aerosol effect on cloud droplet size monitored from  
31 satellite. *Science*, 295, 834-838.
- 32 Breshears, D.D., and C.D. Allen, 2002: The importance of rapid, disturbance-induced losses in carbon  
33 management and sequestration. *Global Ecology & Biogeography*, 11, 1-5.
- 34 Brewer, P.G., E.T. Peltzer, G. Friedrich, I. Aya, and K. Amame, 2000: Experiments on the ocean  
35 sequestration of fossil fuel CO<sub>2</sub>: pH measurements and hydrate formation. *Marine Chemistry*, 72(2-4),  
36 83-93.
- 37 Broecker, W.S., and T. Takahashi, 1978: Neutralization of fossil fuel CO<sub>2</sub> by marine calcium carbonate. In:  
38 *The fate of fossil fuel CO<sub>2</sub> in the ocean* [Andersen, N.R. and A. Malahoff (eds.)]. Plenum Press, New  
39 York, pp. 213-248.
- 40 Broecker, W.S., and T.-H. Peng, 1982: *Tracers in the sea*. ELDIGIO Press, New York, 689 pp.
- 41 Broecker, W.S., and T.-H. Peng, 1986: Carbon cycle: 1985 – glacial to interglacial changes in the operation  
42 of the global carbon cycle. *Radiocarbon*, 28, 309-327.
- 43 Broerse, A.T.C., T. Tyrrell, J.R. Young, A.J. Poulton, A. Merico, W.M. Balch, P.I. Miller, 2003: The cause  
44 of bright waters in the Bering Sea in winter. *Continental Shelf Research*, 23, 1579-1596.
- 45 Brook, E., S. Harder, J. Severinghaus, E. Steig, and C. Sucher C, 2000: On the origin and timing of rapid  
46 changes in atmospheric methane during the last glacial period. *Global Biogeochemical Cycles*, 14, 559-  
47 572.
- 48 Brovkin, V., A. Ganopolski, M. Claussen, C. Kubatzki, V. Petoukhov, 1999: Modelling climate response to  
49 historical land cover change. *Global Ecology and Biogeography*, 8(6), 509–517.
- 50 Brovkin, V., S. Sitch, W. von Bloh, M. Claussen, E. Bauer, and W. Cramer, 2004: Role of land cover  
51 changes for atmospheric CO<sub>2</sub> increase and climate change during the last 150 years. *Global Change  
52 Biology*, 10, 1253–1266, doi:10.1111/j.1365-2486.2004.00812.
- 53 Browell, E.V., J.W. Hair, C.F. Butler, W.B. Grant, R.J. DeYoung, M.A. Fenn, V.G. Brackett, M.B. Clayton,  
54 L.A. Brasseur, D.B. Harper, B.A. Ridley, A.A. Klonecki, P.G. Hess, L.K. Emmons, X.X. Tie, E.L  
55 Atlas, C.A. Cantrell, A.J. Wimmers, D.R. Blake, M.T. Coffey, J.W. Hannigan, J.E. Dibb, R.W. Talbot,  
56 F. Flocke, A.J. Weinheimer, A. Fried, B. Wert, J.A. Snow, B.L. Lefer, 2003: Ozone, aerosol, potential

- 1 vorticity, and trace gas trends observed at high-latitudes over North America from February to May  
2 2000. *Journal of Geophysical Research*, 108(D4), 8369, doi: 10.1029/2001JD001390.
- 3 Brown, S., and A.E. Lugo, 1982: The storage and production of organic-matter in tropical forests and their  
4 role in the global carbon-cycle. *Biotropica*, 14(3), 161-187.
- 5 Brown, T.J., B.L. Hall, and A.L. Westerling, 2004: The impact of twenty-first century climate change on  
6 wildland fire danger in the western United States: an applications perspective. *Climatic Change*, 62,  
7 365-388.
- 8 Brunner, D., J. Staehelin, H.L. Rogers, M.O. Kohler, J.A. Pyle, D. Hauglustaine, L. Jourdain, T.K. Berntsen,  
9 M. Gauss, I.S.A. Isaksen, E. Meijer, P. van Velthoven, G. Pitari, E. Mancini, V. Grewe, R. Sausen,  
10 2003: An evaluation of the performance of chemistry transport models by comparison with research  
11 aircraft observations. Part 1: concepts and overall model performance. *Atmospheric Chemistry &*  
12 *Physics*, 3, 1609-1631.
- 13 Buermann, W., Y. Wang, J. Dong, L. Zhou, Z. Zeng, R.E. Dickinson, C.S. Potter, and R.B. Myneni, 2002:  
14 Analysis of a multiyear global vegetation leaf area index data set. *Journal of Geophysical Research*,  
15 107(D22), 4646, doi:10.1029/2001JD000975.
- 16 Buesseler, K.O., J.E. Andrews, S.M. Pike, and M.A. Charette, 2004: The effects of iron fertilization on  
17 carbon sequestration in the Southern Ocean. *Science*, 304, 414-417.
- 18 Buffett, B., and D. Archer, 2004: Global inventory of methane clathrate: sensitivity to changes in the deep  
19 ocean. *Earth and Planetary Sciences Letters*, 227, 185-199.
- 20 Burkhardt, S., I. Zondervan, and U. Riebesell, 1999: Effect of CO<sub>2</sub> concentration on C:N:P ratio in marine  
21 phytoplankton: a species comparison. *Limnology and Oceanography*, 44(3), 683-690.
- 22 Byrne, R.H., and D.R. Kester, 1976: Solubility of hydrous ferric oxide and iron speciation in sea water.  
23 *Marine Chemistry*, 4, 225-274.
- 24 Caldeira, K., and M.E. Wickett, 2003: Anthropogenic carbon and ocean pH. *Nature*, 425(6956), 365-368.
- 25 Caldeira, K., M. Akai, P. Brewer, B. Chen, P. Haugan, T. Iwama, P. Johnston, H. Kheshgi, Q. Li, T.  
26 Ohsumi, H. Pörtner, C. Sabine, Y. Shirayama, J. Thomson, 2005: Chapter 6: Ocean storage. In: *IPCC*  
27 *Special Report on Carbon Dioxide Capture and Storage*. (Available from <http://www.ipcc.ch> after  
28 September 26<sup>th</sup>, 2005).
- 29 Cao, M., Gregson K. and Marshall S. 1998. Global methane emission from wetlands and its sensitivity to  
30 climate change. *Atmospheric Environment*, 32, 3291-3299.
- 31 Capone, D.G., J.P. Zehr, H.W. Paerl, B. Bergman, and E.J. Carpenter, 1997: *Trichodesmium*: a globally  
32 significant marine cyanobacterium. *Science*, 276, 1221-1229.
- 33 Cardoso, M.F., G.C. Hurtt, B. Moore III, C.A. Nobre, and E. Prins, 2003: Projecting future fire activity in  
34 Amazônia. *Global Change Biology*, 9, 656-669.
- 35 Carlson, C.A., H.W. Ducklow, and A.F. Michaels, 1994: Annual flux of dissolved organic carbon from the  
36 euphotic zone in the northwestern Sargasso Sea. *Nature*, 371, 405-408.
- 37 Cattanio, J.H., E.A. Davidson, D.C. Nepstad, L.V. Verchot, and I.L. Ackerman, 2002: Unexpected results of  
38 a pilot throughfall exclusion experiment on soil emissions of CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, and NO in eastern  
39 Amazonia. *Biological Fertility of Soils*, 36, 102-108.
- 40 Chagnon, F.J.F., R.L. Bras, and J. Wang, 2004: Climatic shift in patterns of shallow clouds over the  
41 Amazon. *Geophysical Research Letters*, 31(24), L24212, doi:10.1029/2004GL021188.
- 42 Chambers, J.Q., and S.E. Trumbore, 1999: An age-old problem. *Trends in Plant Science*, 4(10), 385-386.
- 43 Chambers, J.Q., N. Higuchi, L.M. Teixeira, J. dos Santos, S.G. Laurance, and S.E. Trumbore, 2004a:  
44 Response of tree biomass and wood litter to disturbance in a Central Amazon forest. *Oecologia*, 141(4),  
45 596-611.
- 46 Chambers, J.Q., E.S. Tribuzy, L.C. Toledo, B.F. Crispim, N. Higuchi, J. dos Santos, A.C. Araujo, B. Kruijt,  
47 A.D. Nobre, and S.E. Trumbore, 2004b: Respiration from a tropical forest ecosystem: Partitioning of  
48 sources and low carbon use efficiency. *Ecological Applications*, 14(4), S72-S88.
- 49 Chambers, S.D., J. Beringer, J.T. Randerson, and F.S. Chapin, 2005: Fire effects on net radiation and energy  
50 partitioning: Contrasting responses of tundra and boreal forest ecosystems. *Journal of Geophysical*  
51 *Research*, 110(D9), D09106, doi: 10.1029/2004JD005299.
- 52 Chameidis, W.L., C. Lou, R. Saylor, D. Streets, Y. Huang, M. Bergin, and F. Giorgi, 2002: Correlation  
53 between model-calculated anthropogenic aerosols and satellite-derived cloud optical depths: indication  
54 of indirect effect? *Journal of Geophysical Research*, 107, doi:10.1029/JD000208.
- 55 Chance, K., P. Palmer, R.J.D. Spurr, R.V. Martin, T. Kurosu, and D.J. Jacob, 2000: *Satellite observations of*  
56 *formaldehyde over North America from GOME*. *Geophysical Research Letters*, 27, 3461-3464.

- 1 Chandra, S., J.R. Ziemke, and R.V. Martin, 2003: Tropospheric ozone at tropical and middle latitudes  
2 derived from TOMS/MLS residual: comparison with a global model. *Journal of Geophysical Research*,  
3 108(D9), 4291, doi:10.1029/2002JD002912.
- 4 Chapela, I.H., L.J. Osher, T.R. Horton, and M.R. Henn, 2001: Ectomycorrhizal fungi introduced with exotic  
5 pine plantations induce soil carbon depletion. *Soil Biology & Biochemistry*, 33, 1733-1740.
- 6 Chapman, S.J., and M. Thurlow, 1996: The influence of climate on CO<sub>2</sub> and CH<sub>4</sub> emissions from organic  
7 soils. *Journal of Agricultural and Forest Meteorology*, 79, 205-217.
- 8 Charlson, R.J., J.E. Lovelock, M.O. Andreae, and S.G. Warren, 1987: Oceanic phytoplankton, atmospheric  
9 sulphur, cloud albedo, and climate. *Nature*, 326, 655-661.
- 10 Chave, J., R. Condit, S. Lao, J.P. Caspersen, R.B. Foster, and S.P. Hubbell, 2003b: Spatial and temporal  
11 variation of biomass in a tropical forest: results from a large census plot in Panama. *Journal of Ecology*,  
12 91, 240-252.
- 13 Chen, M., P. Xie, and J.E. Janowiak, 2002: Global land precipitation: a 50-yr monthly analysis based on  
14 gauge observations. *Journal of Hydrometeorology*, 3, 249-266.
- 15 Chen, Y-H., and R.G. Prinn, 2005a: Atmospheric modeling of high- and low-frequency methane  
16 observations: importance of interannually varying transport, (in press).
- 17 Chen, Y-H., and R.G. Prinn, 2005b: Estimation of atmospheric methane emission between 1996-2001 using  
18 a 3-D global chemical transport model, (submitted).
- 19 Cheng, Y., U. Lohmann, J. Zhang, Y. Luo, Z. Liu, and G. Lesins, 2005: Contribution of changes in sea  
20 surface temperature and aerosol loading to the decreasing precipitation trend in Southern China. *Journal*  
21 *of Climate*, 18(9), 1381-1390.
- 22 Chiappello, I., C. Moulin, and J.M. Prospero, 2005: Understanding the long-term variability of African dust  
23 transport across the Atlantic as recorded in both Barbados surface concentrations and large-scale Total  
24 Ozone Mapping Spectrometer (TOMS) optical thickness. *Journal of Geophysical Research-*  
25 *Atmospheres*, 110, doi:10.1029/2004JD005132.
- 26 Chisholm, S.W., P.G. Falkowski, and J.J. Cullen, 2001: Dis-crediting ocean fertilization. *Science*, 294, 309-  
27 310.
- 28 Chou, M.D., P.K. Chan, and M.H. Wang, 2002: Aerosol radiative forcing derived from SeaWiFS-retrieved  
29 aerosol optical properties. *Journal of Atmospheric Science*, 59, 748-757.
- 30 Choudhury, B.J., 2000: A sensitivity analysis of the radiation use efficiency for gross photosynthesis and net  
31 carbon accumulation by wheat. *Journal of Agricultural and Forest Meteorology*, 101, 217-234.
- 32 Christensen, T.R., A. Ekberg, L. Ström, and M. Mastepanov, 2003: Factors controlling large scale variations  
33 in methane emission from wetlands. *Geophysical Research Letters*, 30, 1414,  
34 doi:10.1029/2002GL016848.
- 35 Chung, C.E., and G.J. Zhang, 2004: Impact of absorbing aerosol on precipitation: dynamic aspects in  
36 association with convective available potential energy and convective parameterization closure and  
37 dependence on aerosol heating profile. *Journal of Geophysical Research*, 109,  
38 doi:10.1029/2004JD004726.
- 39 Chung, C.E., V. Ramanathan and J.T. Kiehl, 2002: Effects of South Asian absorbing haze on the Northeast  
40 monsoon and surface-air heat exchange. *Journal of Climate*, 15, 2462-2476.
- 41 Cicerone, R., J. Orr, P. Brewer, P.Haugan, L. Merlivat, T. Ohsumi, S. Pantoja, and. H.-O. Poertner, 2004:  
42 The ocean in a high CO<sub>2</sub> world. *EOS*, 85(37), 351- 353.
- 43 Clair, T.A., J.M. Ehrman, and K. Higuchi, 1999: Changes in freshwater carbon exports from Canadian  
44 terrestrial basins to lakes and estuaries under 2xCO<sub>2</sub> atmospheric scenario. *Global Biogeochemical*  
45 *Cycles*, 13(4), 1091-1097.
- 46 Clark, D.A., 2004a: Sources or sinks? The responses of tropical forests to current and future climate and  
47 atmospheric composition. *Philosophical Transactions of the Royal Society of London, Series B*, 359,  
48 477-491.
- 49 Clark, D.A., 2004b: Tropical forests and global warming: slowing it down or speeding it up? *Frontiers in*  
50 *Ecology & the Environment*, 2(2), 73-80.
- 51 Clark, D.A., S.C. Piper, C.D. Keeling, and D.B. Clark, 2003: Tropical rain forest tree growth and  
52 atmospheric carbon dynamics linked to interannual temperature variation during 1984-2000.  
53 *Proceedings of the National Academy of Sciences USA*, 100, 5852-5857.
- 54 Clark, D.B., C.M. Taylor, and A.J. Thorpe, 2004: Feedback between the land surface and rainfall at  
55 convective length scales. *Journal of Hydrometeorology*, 5(4), 625-639.
- 56 Coakley Jr., J.A., and C.D. Walsh, 2002: Limits to the aerosol indirect radiative forcing derived from  
57 observations of ship tracks. *Journal of Atmospheric Science*, 59, 668-680.

- 1 Cochrane, M.A., 2003: Fire science for rainforests. *Nature*, 421(6926), 913-919.
- 2 Cochrane, M.A., and W.F. Laurance, 2002: Fire as a large-scale edge effect in Amazonian forests. *Journal of*  
3 *Tropical Ecology*, 18, 311-325.
- 4 Cohan, D.S., J. Xu, R. Greenwald, M.H. Bergin, and W.L. Chameides, 2002: Impact of atmospheric aerosol  
5 light scattering and absorption on terrestrial net primary productivity. *Global Biogeochemical Cycles*,  
6 16(4), 25-34, 1090, doi:10.1029/2001GB001441.
- 7 Collatz, G.J., L. Bounoua, S.O. Los, D.A. Randall, I.Y. Fung, and P.J. Sellers, 2000: A mechanism for the  
8 influence of vegetation on the response of the diurnal temperature range to changing climate.  
9 *Geophysical Research Letters*, 27(20), 3381-3384.
- 10 Collier, J.C., and K.P. Bowman, 2004: Diurnal cycle of tropical precipitation in a general circulation model.  
11 *Journal of Geophysical Research*, 109, D17105, doi:10.1029/2004JD004818.
- 12 Collins, W.J., R.G. Derwent, B. Garnier, C.E. Johnson, M.G. Sanderson, and D.S. Stevenson, 2003: Effect  
13 of stratosphere-troposphere exchange on the future tropospheric ozone trend. *Journal of Geophysical*  
14 *Research*, 108(D12), 8528, doi:10.1029/2002JD002617.
- 15 Conrad, R., 1996: Soil microorganisms as controllers of atmospheric trace gases (H<sub>2</sub>, CO, CH<sub>4</sub>, OCS, N<sub>2</sub>O,  
16 and NO). *Microbiology Review*, 60, 609-640.
- 17 Conrad, R., and W. Seiler, 1980: Contribution of hydrogen production by biological nitrogen fixation to the  
18 global hydrogen budget. *Journal of Geophysical Research*, 85, 5493-5498.
- 19 Constable, J.V.H., A.B. Guenther, D.S. Schimel, and R.K. Monson, 1999: Modeling changes in VOC  
20 emission in response to climate change in the continental United States. *Global Change Biology*, 5, 791-  
21 806.
- 22 Conway, T.J., and P. Tans, 2004: Atmospheric CO<sub>2</sub> records from sites in the CMDL air sampling network.  
23 In: *Trends Online: A Compendium of Data on Global Change*. Carbon Dioxide Information Analysis  
24 Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tenn., U.S.A.
- 25 Cooke, W.F., and J.J.N. Wilson, 1996: A global black carbon aerosol model. *Journal of Geophysical*  
26 *Research*, 101, 19395-19409.
- 27 Coops, N.C., and R.H. Waring, 2001: Assessing forest growth across southwestern Oregon under a range of  
28 current and future global change scenarios using a process model, 3-PG. *Global Change Biology*, 7(1),  
29 15-29.
- 30 Coops, N.C., R.H. Waring, and J.J. Landsberg, 2001: Estimation of potential forest productivity across the  
31 Oregon transect using satellite data and monthly weather records. *International Journal of Remote*  
32 *Sensing*, 22(18), 3797-3812.
- 33 Cox, P.M., R.A. Betts, C.D. Jones, S.A. Spall, and I.J. Totterdell, 2000: Acceleration of global warming due  
34 to carbon-cycle feedbacks in a coupled climate model. *Nature*, 408(6809), doi:10.1038/35041539.
- 35 Cox, P.M., R.A. Betts, M. Collins, C. Harris, C. Huntingford, C.D. Jones, 2004: Amazonian forest dieback  
36 under climate-carbon cycle projections for the 21st century. *Theoretical and Applied Climatology*, 78,  
37 137-156.
- 38 Cramer, W., A. Bondeau, F.I. Woodward, I.C. Prentice, R.A. Betts, V. Brovkin, P.M. Cox, V. Fisher, J.A.  
39 Foley, A.D. Friend, C. Kucharik, M.R. Lomas, N. Ramankutty, S. Sitch, B. Smith, A. White, and C.  
40 Young-Molling, 2001: Global response of terrestrial ecosystem structure and function to CO<sub>2</sub> and  
41 climate change: results from six dynamic global vegetation models. *Global Change Biology*, 7(4), 357-  
42 374.
- 43 Cramer, W., A. Bondeau, S. Schaphoff, W. Lucht, B. Smith, and S. Sitch, 2004: Tropical forests and the  
44 global carbon cycle: impacts of atmospheric carbon dioxide, climate change and rate of deforestation.  
45 *Philosophical Transactions of the Royal Society of London Series B-Biological Sciences*, 359(1443),  
46 331-343.
- 47 Creilson, J.K., J. Fishman, and A.E. Wozniak, 2003: Intercontinental transport of tropospheric ozone: a  
48 study of its seasonal variability across the North Atlantic using tropospheric ozone residuals and its  
49 relationship to the North Atlantic Oscillation. *Atmospheric Chemistry & Physics*, 3, 2053-2066.
- 50 Croft, B., U. Lohmann, and K. Von Salzen, 2005: Black carbon ageing in the Canadian Centre for Climate  
51 modelling and analysis atmospheric general circulation model. *Atmospheric Chemistry & Physics*  
52 *Discussions*, 5, 1383-1419.
- 53 Crowley, T.J., 1990: Are there any satisfactory geologic analogs for a future greenhouse warming. *Journal*  
54 *of Climate*, 3(11), 1282-1293.
- 55 Crucifix, M., R.A. Betts, and P.M. Cox, 2005: Vegetation and climate variability: a GCM modeling study.  
56 *Climate Dynamics*, 24, 457-467, doi:10.1007/S00382-004-0504-z.

- 1 Cunnold, D.M., L.P. Steele, P.J. Fraser, P.G. Simmonds, R.G. Prinn, R.F. Weiss, L.W. Porter, S. O'Doherty,  
2 R.L. Langenfelds, P.B. Krummel, H.J. Wang, L. Emmons, X.X. Tie, and E.J. Dlugokencky, 2002: In  
3 situ measurements of atmospheric methane at GAGE/AGAGE sites during 1985-2000 and resulting  
4 source inferences. *Journal of Geophysical Research*, 107(D14), 4225, doi:10.1029/2001JD001226.
- 5 Curran, L.M., S.N. Trigg, A.K. McDonald, D. Astiani, Y.M. Hardiono, P. Siregar, I. Caniago, and E.  
6 Kasischke, 2004: Lowland forest loss in protected areas of Indonesian Borneo. *Science*, 303(5660),  
7 1000-1003.
- 8 Cziczo, D.J., P.J. DeMott, S.D. Brooks, A.J. Prenni, D.S. Thomson, D. Baumgardner, J.C. Wilson, S.M.  
9 Kreidenweis, and D.M. Murphy, 2004: Observations of organic species and atmospheric ice formation.  
10 *Geophysical Research Letters*, 31, doi:10.1029/2004GL019822.
- 11 Czikowsky, M.J., and D.R. Fitzjarrald, 2004: Evidence of seasonal changes in evapotranspiration in eastern  
12 U.S. hydrological records. *Journal of Hydrometeorology*, 5, 974-988.
- 13 D'Odorico, P., and A. Porporato, 2004: Preferential states in soil moisture and climate dynamics.  
14 *Proceedings of the National Academy of Sciences of the USA*, 101(24), 8848-8851.
- 15 Dai, A., and K.E. Trenberth, 2002: Estimates of freshwater discharge from continents: latitudinal and  
16 seasonal variations. *Journal of Hydrometeorology*, 3, 660-687.
- 17 Dai, Y., X. Zeng, R.E. Dickinson, I. Baker, G.B. Bonan, M.G. Bosilovich, A.S. Denning, P.A. Dirmeyer,  
18 P.R. Houser, G.-Y. Niu, K.W. Oleson, C.A. Schlosser, and Z.-L. Yang, 2003: The Common Land Model  
19 (CLM). *Bulletin of the American Meteorological Society*, 84(8), 1013-1023.
- 20 Dai, Y., R.E. Dickinson, and Y.-P. Wang, 2004: A two-big-leaf model for canopy temperature,  
21 photosynthesis and stomatal conductance. *Journal of Climate*, 17, 2281-2299.
- 22 Daniel, J. S. and S. Solomon, 1998: On the climate forcing of carbon monoxide. *Journal of Geophysical*  
23 *Research-Atmospheres*, 103(D11), 13249-13260.
- 24 Dargaville, R.J., S.C. Doney, and I.Y. Fung, 2003: Inter-annual variability in the interhemispheric  
25 atmospheric CO<sub>2</sub> gradient: contributions from transport and the seasonal rectifier. *Tellus Series B-*  
26 *Chemical And Physical Meteorology*, 55(2), 711-722.
- 27 Davidson, E.A., F.Y. Ishida, and D.C. Nepstad, 2004: Effects of an experimental drought on soil emissions  
28 of carbon dioxide, methane, nitrous oxide, and nitric oxide in a moist tropical forest. *Global Change*  
29 *Biology*, 10(5), 718-730.
- 30 Davidson, E.A., S.E. Trumbore, and R. Amundson, 2000: Biogeochemistry - soil warming and organic  
31 carbon content. *Nature*, 408(6814), 789-790.
- 32 De Baar, H.J.W., 1992: Options for enhancing the storage of carbon dioxide in the oceans: a review. *Energy*  
33 *conversion and Management*, 33(5-8), 635-642.
- 34 De Baar, H.J.W., P.W. Boyd, K.H. Coale, M.R. Landry, A. Tsuda, P. Assmy, D.C.E. Bakker, Y. Bozec, R.T.  
35 Barber, M.A. Brzezinski, K.O. Buesseler, M. Boyé, P.L. Croot, F. Gervais, M.Y. Gorbunov, P.J.  
36 Harrison, W.T. Hiscock, P.Laan, C. Lancelot, M. Levasseur, A. Marchetti, F.J. Millero, J. Nishioka, Y.  
37 Nojiri, T. van Ojen, U. Riebesell, M.J.A. Rijkenberg, H. Saito, S. Takeda, K.R. Timmermanns, and  
38 M.J.W. Veldhuis, 2005: Synthesis of 8 iron fertilization experiments: from the iron age in the age of  
39 enlightenment. *Journal of Geophysical Research (Oceans)*, Spec. Vol. The Oceans in a High-CO<sub>2</sub>  
40 World, (accepted).
- 41 de Camargo, P.B., S.E. Trumbore, L.A. Martinelli, E.A. Davidson, D.C. Nepstad, and R.L. Victoria, 1999:  
42 Soil carbon dynamics in regrowing forest of eastern Amazonia. *Global Change Biology*, 5(6), 693-702.
- 43 De Garidel-Thoron, L. Beaufort, F. Bassinot, and P. Henry, 2004: Evidence for large methane release to the  
44 atmosphere from deep-sea gas-hydrate dissociation during the last glacial episode. *Proceedings of*  
45 *National Academy of Science (USA)*, 101, 9187-9192.
- 46 De Leeuw, G., L. Cohen, L.M. Frohn, G. Geernaert, O. Hertel, B. Jensen, T. Jickells, L. Klein, G.J. Kunz, S.  
47 Lund, M. Moerman, F. Muller, B. Pedersen, K. von Salzen, K.H. Schlunzen, M. Schulz, C.A. Skjoth,  
48 L.L. Sorensen, L. Spokes, S. Tamm, and E. Vignati, 2001: Atmospheric input of nitrogen into the North  
49 Sea: ANICE project overview. *Continental Shelf Research*, 21(18-19), 2073-2094.
- 50 DeLucia, E.H., D.J. Moore, and R.J. Norby, 2005: Contrasting responses of forest ecosystems to rising  
51 atmospheric CO<sub>2</sub>: implications for the global C cycle. *Global Biogeochemical Cycles*, 19, G3006, doi:  
52 10.1029/2004GB002346
- 53 De Moura, M.L., and L.S. Galvao, 2003: Smoke effects on NDVI determination of savannah vegetation  
54 types. *International Journal of Remote Sensing*, 24(21), 4225-4231.
- 55 de Rosnay, P., J. Polcher, M. Bruen, and K. Laval, 2002: Impact of a physically based soil water flow and  
56 soil-plant interaction representation for modeling large-scale land surface processes. *Journal of*  
57 *Geophysical Research*, 107(D11), 4118, doi:10.1029/2001JD000634.

- 1 Decesari, S., M.C. Facchini, M. Mircea, F. Cavalli, and S. Fuzzi, 2003: Solubility properties of surfactants in  
2 atmospheric aerosol and cloud/fog water samples. *Journal of Geophysical Research*, 108,  
3 doi:10.1029/2003JD003566.
- 4 DeFries, R.S., R.A. Houghton, M.C. Hansen, C.B. Field, D. Skole, and J. Townshend, 2002a: Carbon  
5 emissions from tropical deforestation and regrowth based on satellite observations for the 1980s and  
6 1990s. *Proceedings of the National Academy of Sciences (USA)*, 99, 14256–14261.
- 7 DeFries, R.S., L. Bounoua, and G.J. Collatz, 2002b: Human modification of the landscape and surface  
8 climate in the next fifty years. *Global Change Biology*, 8(5), 438–458.
- 9 Del Grosso, S.J., W.J. Parton, A.R. Mosier, D.S. Ojima, C.S. Potter, W. Boroken, R. Brumme. K. Butterbach-  
10 Bahl, P.M. Crill, K. Dobbie, and K.A. Smith, 2000: General CH<sub>4</sub> oxidation model and comparison of  
11 CH<sub>4</sub> oxidation in natural and managed systems. *Global Biogeochemical Cycles*, 14, 999–1019.
- 12 Del Grosso, S.J., A.R. Mosier, W.J. Parton, and D.S. Ojima, 2005: DAYCENT model analysis of past and  
13 contemporary soil N<sub>2</sub>O and net greenhouse gas flux for major crops in the USA. *Soil & Tillage  
14 Research*, 83(1), 9–24.
- 15 Denning, A., M. Holzer, K. Gurney, M. Heimann, R. Law, P. Rayner, I. Fung, S.-M. Fan, S. Taguchi, P.  
16 Friedlingstein, Y. Balkanski, J. Taylor, M. Maiss, and I. Levin, 1999: Three-dimensional transport and  
17 concentration of SF<sub>6</sub>: a model intercomparison study (Transcom 2). *Tellus B*, 51, 266–297.
- 18 Dentener, F., W. Peters, M. Krol, M. van Weele, P. Bergamaschi, and J. Lelieveld, 2003a: Interannual  
19 variability and trend of CH<sub>4</sub> lifetime as a measure for OH changes in the 1979–1993 time period.  
20 *Journal of Geophysical Research-Atmospheres*, 108(D15), 4442, doi:10.1029/2002JD002916.
- 21 Dentener, F., M. van Weele, M. Krol, S. Houweling, and P. van Velthoven, 2003b: Trends and inter-annual  
22 variability of methane emissions derived from 1979–1993 global CTM simulations. *Atmospheric  
23 Chemistry & Physics*, 3, 73–88.
- 24 Dentener, F., D. Stevenson, J. Cofala, R. Mechler, M. Amann, P. Bergamaschi, F. Raes, and R. Derwent,  
25 2004: The impact of air pollutant and methane emission controls on tropospheric ozone and radiative  
26 forcing: CTM calculations for the period 1990–2030. *Atmospheric Chemistry & Physics Discussions*, 4,  
27 1–68.
- 28 Dentener, F., D. Stevenson, K. Ellingsen, T. van Noije, M. Schultz, M. Amann, C. Atherton, N. Bell, D.  
29 Bergmann, I. Bey, L. Bouwman, T. Butler, J. Cofala, B. Collins, R. Doherty, J. Drevet, B. Eickhout, H.  
30 Eskes, A. Fiore, M. Gauss, D. Hauglustaine, L. Horowitz, I. Isaksen, B. Josse, M. Krol, J.F. Müller,  
31 V.H. Peuch, G. Pitari, J. Pyle, S. Rast, J. Rodriguez, M. Sanderson, N. Savage, D. Shindell, S. Strahan,  
32 K. Sudo, S. Szopa, R. van Dingenen, O. Wild, and G. Zeng, 2005: Global air quality for the next  
33 generation. *Geophysical Research Letters*, (submitted).
- 34 Derwent, R.G., W.J. Collins, C.E. Johnson, and D.S. Stevenson, 2001a: Transient behaviour of tropospheric  
35 ozone precursors in a global 3-D CTM and their indirect greenhouse effects. *Climatic Change*, 49(4),  
36 463–487.
- 37 Desborough, C.E., 1999: Surface energy balance complexity in GCM land surface models. *Climate  
38 Dynamics*, 15, 389–403.
- 39 Dickens, G.R., 2001: Modeling the global carbon cycle with gas hydrate capacitor: Significance for the latest  
40 Paleocene thermal maximum. In: *Natural Gas Hydrates: Occurrence, Distribution, and Detection*  
41 [Paull, C.K. and W.P. Dillon (eds.)]. *Geophysical Monograph*, 124, American Geophysical Union,  
42 Washington, DC, 19–38.
- 43 Dickinson, R.E., J.A. Berry, G.B. Bonan, G.J. Collatz, C.B. Field, I.Y. Fung, M. Goulden, W.A. Hoffman,  
44 R.B. Jackson, R. Myneni, P.J. Sellers, and M. Shaikh, 2002: Nitrogen controls on climate model  
45 evapotranspiration. *Journal of Climate*, 15(3), 278–295.
- 46 Dickinson, R.E., G. Wang, X. Zeng, and Q.-C. Zeng, 2003: How does the partitioning of evapotranspiration  
47 and runoff between different processes affect the variability and predictability of soil moisture and  
48 precipitation? *Advances in Atmospheric Sciences*, 20(3), 475–478.
- 49 Dickinson, R., K.W. Oleson, G. Bonan, F. Hoffman, P. Thornton, M. Vertenstein, Z-L. Yang, and X. Zeng,  
50 2005: The community land model and its climate statistics as a component of the community climate  
51 system model. *Journal of Climate*, (submitted).
- 52 Dioumaeva, I., S. Trumbore, E.A.G. Schuur, M.L. Goulden, M. Litvak, and A.I. Hirsch, 2002:  
53 Decomposition of peat from upland boreal forest: Temperature dependence and sources of respired  
54 carbon. *Journal of Geophysical Research-Atmospheres*, 108(D3), 8222, doi:10.1029/2001JD000848.
- 55 Dirmeyer, P.A., 2001: An evaluation of the strength of land-atmosphere coupling. *Journal of  
56 Hydrometeorology*, 2(4), 329–344.

- 1 Dlugokencky, E.J., K.A. Masarie, P.M. Lang, P.P. Tans, L.P. Steele, and E.G. Nisbet, 1994: A dramatic  
2 decrease in the growth rate of atmospheric methane in the northern hemisphere during 1992.  
3 *Geophysical Research Letters*, 21, 45–48.
- 4 Dlugokencky, E.J., E.C. Dutton, P.C. Novelli, P.P. Tans, K.A. Masarie, K.O. Lantz, and S. Madronich,  
5 1996: Changes in CH<sub>4</sub> and CO growth rates after the eruption of Mount Pinatubo and their link with  
6 changes in tropical UV flux. *Geophysical Research Letters*, 23, 2761–2764.
- 7 Dlugokencky, E.J., K.A. Masarie, P.M. Lang, and P.P. Tans, 1998: Continuing decline in the growth rate of  
8 the atmospheric methane burden. *Nature*, 393, 447–450.
- 9 Dlugokencky, E.J., B.P. Walter, K.A. Masarie, P.M. Lang, and E.S. Kasischke, 2001: Measurements of an  
10 anomalous global methane increase during 1998. *Geophysical Research Letters*, 28, 499–502.
- 11 Dlugokencky, E.J., S. Houweling, L. Bruhwiler, K.A. Masarie, P.M. Lang, J.B. Miller, and P.P. Tans, 2003:  
12 Atmospheric methane levels off: Temporary pause or a new steady state. *Geophysical Research Letters*,  
13 30(19), doi:10.1029/2003GL018126.
- 14 Doherty, R.M., D.S. Stevenson, W.J. Collins, and M.G. Sanderson, 2005: Influence of convective transport  
15 on tropospheric ozone and its precursors in a chemistry-climate model. *Atmospheric Chemistry &*  
16 *Physics*, 5, 3747–3771.
- 17 Doney, S.C., K. Lindsay, K. Caldeira, J.M. Campin, H. Drange, J.C. Dutay, M. Follows, Y. Gao, A.  
18 Gnanadesikan, N. Gruber, A. Ishida, F. Joos, G. Madec, E. Maier-Reimer, J.C. Marshall, R.J. Matear, P.  
19 Monfray, A. Mouchet, R. Najjar, J.C. Orr, G.K. Plattner, J. Sarmiento, R. Schlitzer, R. Slater, I.J.  
20 Totterdell, M.F. Weirig, Y. Yamanaka, and A. Yool, 2004: Evaluating global ocean carbon models: the  
21 importance of realistic physics. *Global Biogeochemical Cycles*, 18(3), GB3017,  
22 doi:10.1029/2003GB002150.
- 23 Doney, S., K. Lindsay, I. Fung, and J. John, 2005: A stable 1000 year coupled climate-carbon cycle  
24 simulation, *J. Climate*, (submitted).
- 25 Douglass, A.R., M.R. Schoeberl, R.B. Rood, and S. Pawson, 2003: Evaluation of transport in the lower  
26 tropical stratosphere in a global chemistry and transport model. *Journal of Geophysical Research*,  
27 108(D9), 4259, doi:10.1029/2002JD002696.
- 28 Duce, R.A., 1995: Sources, distributions and fluxes of mineral aerosols and their relationship to climate. In:  
29 *Aerosol Forcing of Climate* [Charlson, R. J. and J. Heintzenberg (eds.)]. John Wiley & Sons Ltd., pp.  
30 43-72.
- 31 Dufresne, J.L., P. Friedlingstein, M. Berthelot, L. Bopp, P. Ciais, L. Fairhead, H. Le Treut, and P. Monfray,  
32 2002: On the magnitude of positive feedback between future climate change and the carbon cycle.  
33 *Geophysical Research Letters*, 29(10), 1405, doi:10.1029/2001GL013777.
- 34 Dunn, A.L, C.C. Barford, S.C. Wofsy, M.L. Goulden, B.C. Daube, 2005: A long-term record of carbon  
35 exchange in a boreal black spruce forest: means, responses to interannual variability, and decadal  
36 trends. *Global Change Biology*, (submitted).
- 37 Dupre, B., C. Dessert, P. Oliva, Y. Godderis, J. Viers, L. Francois, R. Millot, and J. Gaillardet, 2003: Rivers,  
38 chemical weathering and Earth's climate. *Compte Rendus Geoscience*, 335(16), 1141-1160.
- 39 Durre, I., and J.M. Wallace, 2001: The warm season dip in diurnal temperature range over the eastern United  
40 States. *Journal of Climate*, 14(3), 354-360.
- 41 Dutay, J.C., J.L. Bullister, S.C. Doney, J.C. Orr, R. Najjar, K. Caldeira, J.M. Campin, H. Drange, M.  
42 Follows, Y. Gao, N. Gruber, M. Hecht, A. Ishida, F. Joos, K. Lindsay, G. Madec, E. Maier-Reimer, J.C.  
43 Marshall, R. Matear, P. Monfray, A. Mouchet, G.K. Plattner, J. Sarmiento, R. Schlitzer, R. Slater, I.J.  
44 Totterdell, M.F. Weirig, Y. Yamanaka, and A. Yool, 2002: Evaluation of ocean model ventilation with  
45 CFC-11: comparison of 13 global ocean models. *Ocean Modelling*, 4(2), 89-102.
- 46 Easter, R.C., S.J. Ghan, Y. Zhang, R.D. Saylor, E.G. Chapman, N.S. Laulainen, H. Abdul-Razzak, L.R.  
47 Leung, X. Bian and R.A. Zaveri, 2004: MIRAGE: Model description and evaluation of aerosols and  
48 trace gases. *Journal of Geophysical Research*, 109, doi:10.1029/2004JD004571.
- 49 Edwards, D.P., L.K. Emmons, D.A. Hauglustaine, D.A. Chu, J.C. Gille, Y.J. Kaufman, G. Pétron, L.N.  
50 Yurganov, L. Giglio, M.N. Deeter, V. Yudin, D.C. Ziskin, J. Warner, J.-F. Lamarque, G.L. Francis, S.P.  
51 Ho, D. Mao, J. Chen, E.I. Grechko, and J.R. Drummond, 2004: Observations of carbon monoxide and  
52 aerosols from the Terra satellite: Northern Hemisphere variability. *Journal of Geophysical Research*,  
53 109, D24202, doi:10.1029/2004JD004727.
- 54 Ehhalt, D.H., 1999: Gas phase chemistry of the troposphere. In: *Global Aspects of Atmospheric Chemistry*  
55 [Baumgärtl, H., W. Grünbein, and F. Hensel (eds.)]. Dr. Dietrich Steinkopf Verlag, Darmstadt,  
56 Germany, pp. 21-110.

- 1 Ek, M.B., and A.A.M. Holtslag, 2004: Influence of soil moisture on boundary layer cloud development.  
2 *Journal of Hydrometeorology*, 5, 86-99.
- 3 Elvert, M., J. Greinert, E. Suess, and M.J. Whiticar, 2001: Carbon isotopes of biomarkers derived from  
4 methane-oxidizing microbes at hydrate ridge, Cascadia Convergent Margin. In: *Natural Gas Hydrates:  
5 Occurrence, Distribution, and Detection* [Paull, C.K. and W.P. Dillon (eds.)]. *Geophysical Monograph*,  
6 124, American Geophysical Union, Washington, DC, pp. 115-129.
- 7 Enting, I.G., C.M. Trudinger, and R.J. Francey, 1995: A synthesis inversion of the concentration and <sup>13</sup>C of  
8 atmospheric CO<sub>2</sub>. *Tellus B*, 47, 35-52.
- 9 EPICA community members, 2004: Eight glacial cycles from an Antarctic ice core. *Nature*, 429, 623-628.
- 10 Etiope, G., 2004: GEM-Geologic emission of methane, the missing source in atmospheric methane budget.  
11 *Atmospheric Environment*, 38, 3099-3100.
- 12 European Commission, 2003: Ozone-climate interactions. In: *Air pollution research report* [Isaksen, I.S.A.  
13 (eds.)]. Report 81, EUR 20623, Luxembourg.
- 14 Facchini, M.C., M. Mircea, S. Fuzzi, and R.J. Charlson, 1999: Cloud albedo enhancement by surface-active  
15 organic solutes in growing droplets. *Nature*, 401, 257-259.
- 16 Farrell, J.W., and W.L. Prell, 1989: Climatic change and CaCO<sub>3</sub> preservation: an 800,000 year bathymetric  
17 reconstruction from the central equatorial Pacific Ocean. *Paleoceanography*, 4, 447-466.
- 18 Fearnside, P.M., 2000: Global warming and tropical land-use change: greenhouse gas emissions from  
19 biomass burning, decomposition and soils in forest conversion, shifting cultivation and secondary  
20 vegetation. *Climate Change*, 46, 115-158.
- 21 Feddes, R.A., H. Hoff, M. Bruen, T. Dawson, P. deRosnay, P. Dirmeyer, R.B. Jackson, P. Kabat, A.  
22 Kleidon, A. Lilly, and A.J. Pitman, 2001: Modeling root water uptake in hydrological and climate  
23 models. *Bulletin of the American Meteorological Society*, 82(12), 2797-2809.
- 24 Feely, R.A., C.L. Sabine, K. Lee, W. Berelson, J. Kleypas, V.F. Fabry, and F.J. Millero, 2004: Impact of  
25 anthropogenic CO<sub>2</sub> on the CaCO<sub>3</sub> system in the oceans. *Science*, 305, 362-366.
- 26 Feely, R.A., J. Boutin, C.E. Cosca, Y. Dandonneau, J. Etcheto, H.Y. Inoue, M. Ishii, C. Le Quéré, D.J.  
27 Mackey, M. McPhaden, N. Metzl, A. Poisson, and R. Wanninkhof, 2002: Seasonal and interannual  
28 variability of CO<sub>2</sub> in the equatorial Pacific. *Deep-Sea Research II*, 49, 2443-2469.
- 29 Feichter, J., E. Roeckner, U. Lohmann, and B. Liepert, 2004: Nonlinear aspects of the climate response to  
30 greenhouse gas and aerosol forcing. *Journal of Climate*, 17(12), 2384-2398.
- 31 Feingold, G., W.R. Cotton, S.M. Kreidenweis, and J.T. Davis, 1999: The impact of giant cloud condensation  
32 nuclei on drizzle formation in stratocumulus: Implications for cloud radiative properties. *Journal of  
33 Atmospheric Science*, 56, 4100-4117.
- 34 Feingold, G., W.L. Eberhard, D.E. Veron, and M. Previdi, 2003: First measurements of the Twomey indirect  
35 effect using ground-based remote sensors. *Geophysical Research Letters*, 30,  
36 doi:10.1029/2002GL016633.
- 37 Fekete, B.M., C.J. Vorosmarty, and W. Grabs, 2002: High-resolution fields of global runoff combining  
38 observed river discharge and simulated water balances. *Global Biogeochemical Cycles*, 16,  
39 doi:10.1029/1999GB001254.
- 40 Ferek, R.J., D.A. Hegg, P.V. Hobbs, P. Durkee, and K. Nielsen, 1998: Measurements of ship-induced tracks  
41 in clouds off the Washington coast. *Journal of Geophysical Research*, 103, 23199-23206.
- 42 Fioletov, V.E., G.E. Bodeker, A.J. Miller, R.D. McPeters, and R. Stolarski, 2002: Global and zonal total  
43 ozone variations estimated from ground-based and satellite measurements: 1964–2000. *Journal of  
44 Geophysical Research*, 107(D22), 4647, doi:10.1029/2001JD001350.
- 45 Fischer, H., 2001: Imprint of large-scale atmospheric transport patterns on sea-salt records in northern  
46 Greenland ice cores. *Journal of Geophysical Research*, 106, 23977-23984.
- 47 Flannigan, M.D., B.J. Stocks, and B.M. Wotton, 2000: Climate change and forest fires. *Science of the Total  
48 Environment*, 262(3), 221-229.
- 49 Foley, J.A., M.H. Costa, C. Delire, N. Ramankutty, and P. Snyder, 2003: Green Surprise? How terrestrial  
50 ecosystems could affect earth's climate. *Frontiers in Ecology and the Environment*, 1(1), 38-44.
- 51 Forster, P.M.d.F., and S. Solomon, 2003: Observations of a "weekend effect" in diurnal temperature range.  
52 *Proceeding of the National Academy of Sciences USA*, 100, 11225-11230.
- 53 Franck, V.M., K.W. Bruland, D.A. Hutchins, and M.A. Brzezinski, 2003: Iron and zinc effects on silicic acid  
54 and nitrate uptake kinetics in three high-nutrient, low chlorophyll (HNLC) regions. *Marine Ecology  
55 Progress Series*, 252, 15-33.
- 56 Frankenberg, C., J.F. Meirink, M. van Weele, U. Platt, and T. Wagner, 2005: Assessing methane emission  
57 from global space-borne observation. *Science*, 308, 1010-1014.

- 1 Freedman, J.M., D.R. Fitzjarrald, K.E. Moore, and R.K.Sakai, 2001: Boundary layer clouds and vegetation–  
2 atmosphere feedbacks. *Journal of Climate*, 14, 180-197.
- 3 Freeman, C., N. Fenner, N.J. Ostle, H. Kang, D.J. Dorwick, B. Reynolds, M.A. Lock, D. Sleep, S. Hughes,  
4 and J. Hudson, 2004: Export of dissolved organic carbon from peatlands under elevated carbon dioxide  
5 levels. *Nature*, 430, 195-198.
- 6 Freitas, S.R., K.M. Longo, M.A.F. Silva Dias, P.L. Silva Dias, F.S. Recuero, R. Chatfield, E. Prins, and P.  
7 Artaxo, 2005: Monitoring the transport of biomass burning emissions in South America. *Environmental*  
8 *Fluid Mechanics*, 5, 135-167.
- 9 Frew, R., A. Bowie, P. Croot, and S. Pickmere, 2001: Macronutrient and trace-metal geochemistry of an in  
10 situ iron-induced Southern Ocean bloom. *Deep-Sea Research II*, 48(11-12), 2467-2481.
- 11 Friedl, M.A., 2002: Forward and inverse modeling of land surface energy balance using surface temperature  
12 measurements. *Remote Sensing of Environment*, 79, 344-354.
- 13 Friedlingstein, P., L. Bopp, P. Ciais, J.-L. Dufresne, L. Fairhead, H. LeTreut, P. Monfray, and J. Orr, 2001:  
14 Positive feedback between future climate change and the carbon cycle. *Geophysical Research Letters*,  
15 28, 1543-1546, doi:10.1029/2000GL012015.
- 16 Friedlingstein, P., J.-L. Dufresne, P.M. Cox, and P. Rayner, 2003: How positive is the feedback between  
17 climate change and the carbon cycle? *Tellus Series B*, 55(2), 692-700.
- 18 Friedlingstein, P., P. Cox, R. Betts, L. Bopp, W. von Bloh, V. Brovkin, S. Doney, M. Eby, I. Fung, B.  
19 Govindasamy, J. John, C. Jones, F. Joos, T. Kato, M. Kawamiya, W. Knorr, K. Lindsay, H.D.  
20 Matthews, T. Raddatz, P. Rayner, C. Reick, E. Roeckner, K.-G. Schnitzler, R. Schnur, K. Strassmann,  
21 A.J. Weaver, C. Yoshikawa, and N. Zeng, 2005: Climate-carbon cycle feedback analysis: results from  
22 the C4MIP model intercomparison. *Journal of Climate*, (in press).
- 23 Fu, R., and W. Li, 2004: The influence of the land surface on the transition from dry to wet season in  
24 Amazonia. *Theoretical & Applied Climatology*, 78, 97-110.
- 25 Fuglestad, J.S., T.K. Berntsen, I.S.A. Isaksen, H.T. Mao, X.Z. Liang, and W.C. Wang, 1999: Climatic  
26 forcing of nitrogen oxides through changes in tropospheric ozone and methane: global 3D model  
27 studies. *Atmospheric Environment*, 33(6), 961-977.
- 28 Fuhrman, J.A., and D.G. Capone, 1991: Possible biogeochemical consequences of ocean fertilization.  
29 *Limnology and Oceanography*, 36(8), 1951-1959.
- 30 Fung, I., S.C. Doney, K. Lindsay, and J. John, 2005: Evolution of carbon sinks in a changing climate.  
31 *Proceedings of the National Academy of Sciences USA*, 102(32), 11201-11206.
- 32 Fusco, A.C., and J.A. Logan, 2003: Analysis of 1970-1995 trends in tropospheric ozone at northern  
33 hemisphere midlatitudes with the GEOS-CHEM model. *Journal of Geophysical Research*, 108(D15),  
34 4449, doi:10.1029/2002JD002742.
- 35 Gabric, A.J., R. Simo, R.A. Cropp, A.C. Hirst, and J. Dachs, 2004: Modeling estimates of the global  
36 emission of dimethylsulfide under enhanced greenhouse conditions. *Global Biogeochemical Cycles*  
37 18(2), GB2014, doi:10.1029/2003GB002183.
- 38 Gallagher, M.W., E. Nemitz, J.R. Dorsey, D. Fowler, M.A. Sutton, M. Flynn, and J. Duyzer, 2002:  
39 Measurements and parameterizations of small aerosol deposition velocities to grassland, arable crops,  
40 and forest: Influence of surface roughness length on deposition. *Journal of Geophysical Research*,  
41 107(D12), 4154, doi:10.1029/2001JD000817.
- 42 Galloway, J.N., F.J. Dentener, D.G. Capone, E.W. Boyer, R.W. Howarth, S.P. Seitzinger, G.P. Asner, C.C.  
43 Cleveland, P.A. Green, E.A. Holland, D.M. Karl, A.F. Michaels, J.H. Porter, A.R. Townsend, and C.J.  
44 Vorosmarty, 2004: Nitrogen cycles: past, present, and future. *Biogeochemistry*, 70(2), 153-226.
- 45 Ganzeveld, L.N., J. Lelieveld, F.J. Dentener, M.C. Krol, A.J. Bouwman, and G.-J. Roelofs, 2002: Global  
46 soil-biogenic NO<sub>x</sub> emissions and the role of canopy processes. *Journal of Geophysical Research*,  
47 107(D16), 4298, doi:10.1029/2001JD001289.
- 48 Gao, Z., N. Chae, J. Kim, J. Hong, T. Choi, and H. Lee, 2004: Modeling of surface energy partitioning,  
49 surface temperature, and soil wetness in the Tibetan prairie using the Simple Biosphere Model 2 (SiB2).  
50 *Journal of Geophysical Research*, 109, D06102, doi:10.1029/2003JD004089.
- 51 Gauci, V., E. Matthews, N. Dise, B. Walter, D. Koch, G. Granberg, and M. Vile, 2004: Sulphur pollution  
52 suppression of the wetland methane source in the 20th and 21st centuries. *Proceedings of the National*  
53 *Academy of Science USA*, 101, 12583-12587.
- 54 Gaudinski, J.B., S.E. Trumbore, E.A. Davidson, and S.H. Zheng, 2001: Soil carbon cycling in a temperate  
55 forest: radiocarbon-based estimates of residence times, sequestration rates and partitioning of fluxes  
56 (vol 51, pg 33, 2000). *Biogeochemistry*, 52(1), 113-114.

- 1 Gedney, N., and P. Cox, 2003: The sensitivity of global climate model simulations to the representation of  
2 soil moisture heterogeneity. *Journal of Hydrometeorology*, 4, 1265-1275.
- 3 Gerber, S., F. Joos, P.P. Brügger, T.F. Stocker, M.E. Mann, S. Sitch, and M. Scholze, 2003: Constraining  
4 temperature variations over the last millennium by comparing simulated and observed atmospheric  
5 CO<sub>2</sub>. *Climate Dynamics*, 20, 281-299, doi: 10.1007/s00382-002-0270-8.
- 6 Geron, C., A. Guenther, J. Greenberg, H.W. Loescher, D. Clark, and B. Baker, 2002: Biogenic volatile  
7 organic compound emissions from a lowland tropical wet forest in Costa Rica. *Atmospheric*  
8 *Environment*, 36, 3793-3802.
- 9 Gerten, D., S. Schaphoff, U. Haberlandt, W. Lucht, and S. Sitch, 2004: Terrestrial vegetation and water  
10 balance - hydrological evaluation of a dynamic global vegetation model. *Journal of Hydrology*, 286(1-  
11 4), 249-270.
- 12 Ghan, S.J., G. Guzman, and H. Abdul-Razzak, 1998: Competition between sea salt and sulphate particles as  
13 cloud condensation nuclei. *Journal of Atmospheric Science*, 55, 3340-3347.
- 14 Giannini, A., R. Saravanan, and P. Chang, 2003: Oceanic forcing of Sahel rainfall on interannual to  
15 interdecadal time scales. *Science*, 302, 1027-1030.
- 16 Giardina, C., and M. Ryan, 2000. Evidence that decomposition rates of organic carbon in mineral soil do not  
17 vary with temperature. *Nature*, 404, 858-861.
- 18 Gilgen, H., M. Wild, and A. Ohmura, 1998: Means and trends of shortwave incoming radiation at the surface  
19 estimated from global energy balance archive data. *Journal of Climate*, 11, 2042-2061.
- 20 Ginoux, P., M. Chin, I. Tegen, J. Prospero, B. Holben, O. Dubovik, and S.-J. Lin, 2001: Sources and  
21 distributions of dust aerosols simulated with the GOCART model. *Journal of Geophysical Research*,  
22 16, 20255-20274.
- 23 Gong, S.L., and L.A. Barrie, 2003: Simulating the impact of sea salt on global nss sulphate aerosols. *Journal*  
24 *of Geophysical Research*, 108(D16), 4516, doi:10.1029/2002JD003181.
- 25 Goodale, C.L., M.J. Apps, R.A. Birdsey, C.B. Field, L.S. Heath, R.A. Houghton, J.S. Jenkins, G. Kohlmaier,  
26 W.A. Kurz, S. Liu, G.-J. Nabuurs, S. Nilsson, and A. Shvidenko, 2002: Forest carbon sinks in the  
27 northern hemisphere. *Ecological Applications*, 12(3), 891-899.
- 28 Goulden, M.L., S.C. Wofsy, J.W. Harden, S.E. Trumbore, P.M. Crill, S.T. Gower, T. Fries, B.C. Daube, S.-  
29 M. Fan, D.J. Sutton, A. Bazzaz, and J.W. Munger, 1998: Sensitivity of boreal forest carbon balance to  
30 soil thaw. *Science*, 279(5348), 214-217.
- 31 Govindasamy, B., S. Thompson, A. Mirin, M. Wickett, K. Caldeira, and C. Delire, 2005: Increase of carbon  
32 cycle feedback with climate sensitivity: results from a coupled climate and carbon cycle model. *Tellus*,  
33 578, 153-163.
- 34 Green, P.A., C.J. Vörösmarty, M. Meybeck, J.N. Galloway, B.J. Peterson, and E.W. Boyer, 2004: Pre-  
35 industrial and contemporary fluxes of nitrogen through rivers: a global assessment based on typology.  
36 *Biogeochemistry*, 68(1), 71-105.
- 37 Gregg, W.W., M.E. Conkright, P. Ginoux, J.E. O'Reilly, and N.W. Casey, 2003: Ocean primary production  
38 and climate: Global decadal changes. *Geophysical Research Letters*, 30(15), 1809,  
39 doi:10.1029/2003GL016889.
- 40 Grenfell, J.L., D.T. Shindell, and V. Grewe, 2003: Sensitivity studies of oxidative changes in the troposphere  
41 in 2100 using the GISS GCM. *Atmospheric Chemistry & Physics*, 3, 1267-1283.
- 42 Grewe, V., M. Dameris, R. Hein, R. Sausen, and B. Steil, 2001: Future changes of the atmospheric  
43 composition and the impact on climate change. *Tellus B*, 53(2), 103-121.
- 44 Grimm, A.M., 2003: The El niño impact on the summer monsoon in Brazil: regional processes versus  
45 remote influences. *Journal of Climate*, 16, 263-280.
- 46 Gruber, N., 1998: Anthropogenic CO<sub>2</sub> in the Atlantic Ocean. *Global Biogeochemical Cycles*, 12(1), 165-191.
- 47 Gruber, N., N. Bates, and C.D. Keeling, 2002: Interannual variability in the North Atlantic Ocean carbon  
48 sink. *Science*, 298(5602), 2374-2378.
- 49 Gu, L., D. Baldocchi, S.B. Verma, T.A. Black, T. Vesala, E.M. Falge, and P.R. Dowty, 2002: Advantages of  
50 diffuse radiation for terrestrial ecosystem productivity. *Journal of Geophysical Research*, 107(6), 4050,  
51 doi:10.1029/2001JD001242.
- 52 Gu, L., D.D. Baldocchi, S.C. Wofsy, J.W. Munger, J.J. Michalsky, S.P. Urbanski, and T.A. Boden, 2003:  
53 Response of a deciduous forest to the Mt. Pinatubo eruption: enhanced photosynthesis. *Science*,  
54 299(5615), 2035-2038.
- 55 Guenther, A.B., P.R. Zimmerman, P.C. Harley, R.K. Monson, and R. Fall, 1993: Isoprene and monoterpene  
56 emission rate variability - model evaluations and sensitivity analyses. *Journal of Geophysical Research-*  
57 *Atmospheres*, 98(D7), 12609-12617.

- 1 Guenther, A., C.N. Hewitt, D. Erickson, R. Fall, C. Geron, T. Graedel, P. Harley, L. Klinger, M. Lerdau,  
2 W.A. Mckay, T. Pierce, B. Scholes, R. Steinbrecher, R. Tallamraju, J. Taylor, and P. Zimmerman,  
3 1995: A global-model of natural volatile organic-compound emissions. *Journal of Geophysical*  
4 *Research-Atmospheres*, 100(D5), 8873-8892.
- 5 Guenther, A., B. Baugh, G. Brasseur, J. Greenberg, P. Harley, L. Klinger, D. Serca, and L. Vierling, 1999:  
6 Isoprene emission estimates and uncertainties for the Central African EXPRESSO study domain.  
7 *Journal of Geophysical Research-Atmospheres*, 104(D23), 30625-30639.
- 8 Guillevic, P., R.D. Koster, M.J. Suarez, L. Bounoua, G.J. Collatz, S.O. Los, and S.P.P. Mahanama, 2002:  
9 Influence of the interannual variability of vegetation on the surface energy balance-a global sensitivity  
10 study. *Journal of Hydrometeorology*, 3, 617-629.
- 11 Gurney, K.R., R.M. Law, A.S. Denning, P.J. Rayner, D. Baker, P. Bousquet, L. Bruhwiler, Y-H. Chen, P.  
12 Ciais, S. Fan, I.Y. Fung, M. Gloor, M. Heimann, K. Higushi, J. John, E. Kowalczyk, T. Maki, S.  
13 Maksyutov, P. Peylin, M. Prather, B.C. Pak, J. Sarmiento, S. Tagushi, T. Takahashi, and C.-W. Yuen,  
14 2003: TransCom 3 CO<sub>2</sub> inversion intercomparison: 1. Annual mean control results and sensitivity to  
15 transport and prior flux information. *Tellus B*, 55(2), 555-579.
- 16 Gurney, K.R., R.M. Law, A.S. Denning, P.J. Rayner, B.C. Pak, D. Baker, P. Bousquet, L. Bruhwiler, Y.H.  
17 Chen, P. Ciais, I.Y. Fung, M. Heimann, J. John, T. Maki, S. Maksyutov, P. Peylin, M. Prather, and S.  
18 Taguchi, 2004: Transcom 3 inversion intercomparison: model mean results for the estimation of  
19 seasonal carbon sources and sinks. *Global Biogeochemical Cycles*, 18(1), GB1010,  
20 doi:10.1029/2003GB002111.
- 21 Gutowski, Jr., W.J., F.O. Otieno, R.W. Arritt, E.S. Takle, and Z. Pan, 2004: Diagnosis and attribution of a  
22 seasonal precipitation deficit in a U.S. regional climate simulation. *Journal of Hydrometeorology*, 5,  
23 230-242.
- 24 Haag, W., and B. Kärcher, 2004: The impact of aerosols and gravity waves on cirrus clouds at midlatitudes.  
25 *Journal of Geophysical Research*, 109, doi:10.1029/2004JD004579.
- 26 Haake, B., and V. Ittekkot, 1990: The wind-driven biological pump and carbon removal in the ocean.  
27 *Naturwissenschaften*, 77(2), 75-79.
- 28 Hahmann, A.N., 2003: Representing spatial sub-grid precipitation variability in a GCM. *Journal of*  
29 *Hydrometeorology*, 4(5), 891-900.
- 30 Hahmann, A.N., and R.E. Dickinson, 2001: A fine mesh land approach for general circulation models and its  
31 impact on regional climate. *Journal of Climate*, 14, 1634-1646.
- 32 Hallen, S.J., N. Putnam, C.M. Preston, J.C. Detter, D. Rokhsar, P.M. Richardson, and E.F. DeLong, 2004:  
33 Reverse methanogenesis: Testing the hypothesis with environmental genomics. *Science*, 305, 1457-  
34 1462.
- 35 Handisides, G.M., C. Plass-Dülmer, S. Gilge, H. Bingemer, and H. Berresheim, 2003: Hohenpeissenberg  
36 photochemical experiment (HOPE 2000): measurements and photostationary state calculations of OH  
37 and peroxy radicals. *Atmospheric Chemistry & Physics*, 3, 1565-1588.
- 38 Hansen, J., and L. Nazarenko, 2004: Soot climate forcing via snow and ice albedos. *Proceedings of the*  
39 *National Academy of Sciences USA*, 101, 423-428.
- 40 Hansen, J., A. Lacis, D. Rind, and others, 1983: Climate sensitivity: analysis of feedback mechanisms in  
41 climate processes and climate sensitivity. *Geophysical Monograph*, 29, 130-163.
- 42 Hansen, J., M. Sato, and R. Ruedy, 1995: Long-term changes of the diurnal temperature cycle - implications  
43 about mechanisms of global climate-change. *Atmospheric Research*, 37, 175-209.
- 44 Hansen, J., R. Ruedy, M. Sato, R. Reynolds, 1996: Global surface air temperature in 1995: Return to pre-  
45 Pinatubo level. *Geophysical Research Letters*, 23(13), 1665-1668.
- 46 Harden, J.W., S.E. Trumbore, B.J. Stocks, A. Hirsch, S.T. Gower, K.P. O'Neill, and E.S. Kasischke, 2000:  
47 The role of fire in the boreal carbon budget. *Global Change Biology*, 6, 174-184 (Suppl. 1 Dec.).
- 48 Harvey, H.W., 1969: *The chemistry and fertility of sea waters*, Cambridge University Press, 2<sup>nd</sup> ed., 240 pp.
- 49 Hasumi, H., and S. Emori, 2004: K-1 coupled GCM (MIROC) description. K-1 Technical Report No.1.  
50 Center for Climate System Research (CCSR, Univ. of Tokyo), National Institute for Environmental  
51 Studies (NIES), Frontier Research Center for Global Change (FRCGC).
- 52 Hatzianastassiou, N., B. Katsoulis, and I. Vardavas, 2004: Sensitivity analysis of aerosol direct radiative  
53 forcing in ultraviolet-visible wavelengths and consequences for the heat budget. *Tellus Series B*, 56(4),  
54 368-381.
- 55 Hauglustaine, D.A., and G.P. Brasseur, 2001: Evolution of tropospheric ozone under anthropogenic activities  
56 and associated radiative forcing of climate. *Journal of Geophysical Research*, 106(D23), 32337-32360.

- 1 Hauglustaine, D., and D.H. Ehhalt, 2002: A three-dimensional model of molecular hydrogen in the  
2 troposphere. *Journal of Geophysical Research*, 107(D17), doi:10.1029/2001JD001156.
- 3 Hauglustaine, D.A., F. Hourdin, L. Jourdain, M.A. Filiberti, S. Walters, J.-F. Lamarque, and E.A. Holland,  
4 2004: Interactive chemistry in the laboratoire de meteorologie dynamique general circulation model:  
5 description and background tropospheric chemistry. *Journal of Geophysical Research*, 109, D04314,  
6 doi:10.1029/2003JD003957.
- 7 Hauglustaine, D.A., J. Lathière, and G. Folberth, 2005: Future tropospheric ozone simulated with a climate-  
8 chemistry-biosphere model. *Geophysical Research Letters*, (submitted).
- 9 Heald, C.L., D.J. Jacob, D.B.A. Jones, P.I. Palmer, J.A. Logan, D.G. Streets, G.W. Sachse, J.C. Gille, R.N.  
10 Hoffman, and T. Nehrkorn, 2004: Comparative inverse analysis of satellite (MOPITT) and aircraft  
11 (TRACE-P) observations to estimate Asian sources of carbon monoxide. *Journal of Geophysical  
12 Research*, 109(D23), D23306, doi:10.1029/2004JD005185.
- 13 Heard, D.E., L.J. Carpenter, D.J. Creasey, J.R. Hopkins, J.D. Lee, A.C. Lewis, M.J. Pilling, and P.W.  
14 Seakins, 2004: High levels of the hydroxyl radical in the winter urban troposphere. *Geophysical  
15 Research Letters*, 31, L18112, doi:10.1029/2004GL020544.
- 16 Hedges, J.J., 1992: Global biogeochemical cycles: progress and problems. *Marine Chemistry*, 39, 67-93.
- 17 Heinze, C., 2004: Simulating oceanic CaCO<sub>3</sub> export production in the greenhouse. *Geophysical Research  
18 Letters*, 31, L16308, doi:10.1029/2004GL020613.
- 19 Heinze, C., E. Maier-Reimer, and K. Winn, 1991: Glacial pCO<sub>2</sub> reduction by the World Ocean: experiments  
20 with the Hamburg carbon cycle model. *Paleoceanography*, 6(4), 395-430.
- 21 Heinze, C., A. Hupe, E. Maier-Reimer, N. Dittert, and O. Ragueneau, 2003: Sensitivity of the marine  
22 biospheric Si cycle for biogeochemical parameter variations. *Global Biogeochemical Cycles*, 17(3),  
23 1086, doi:10.1029/2002GB001943.
- 24 Hejzlar, J., M. Dubrovsky, J. Buchtele, and M. Ruzicka, 2003: The apparent and potential effects of climate  
25 change on the inferred concentration of dissolved organic matter in a temperate stream (the Malse  
26 River, South Bohemia). *Science of the Total Environment*, 310(1-3), 143-152.
- 27 Henderson-Sellers, A., P. Irannejad, K. McGuffie, and A.J. Pitman, 2003: Predicting land-surface climates -  
28 better skill or moving targets? *Geophysical Research Letters*, 30(14), 1777,  
29 doi:10.1029/2003GL017387.
- 30 Henderson-Sellers, A., K. McGuffie, D. Noone, and P. Irannejad, 2004: Using stable water isotopes to  
31 evaluate basin-scale simulations of surface water budgets. *Journal of Hydrometeorology*, 5(5), 805-822.
- 32 Hendricks, J., B. Kärcher, M. Ponater, and U. Lohmann, 2005: Do aircraft black carbon emissions affect  
33 cirrus clouds on a global scale? *Geophysical Research Letters*, 32, L12814,  
34 doi:10.1029/2005GL022740.
- 35 Herbert, R.A., 1999: Nitrogen cycling in coastal marine ecosystems. *FEMS Microbiology Reviews*, 23(5),  
36 563-590.
- 37 Hess, J.C., C.A. Scott, G.L. Hufford, and M.D. Fleming, 2001: El Niño and its impact on fire weather  
38 conditions in Alaska. *International Journal of Wildland Fire*, 10(1), 1-13.
- 39 Hicke, J.A., G.P. Asner, J.T. Randerson, C. Tucker, S. Los, R. Birdsey, J.C. Jenkins, and C. Field, 2002a:  
40 Trends in North American net primary productivity derived from satellite observations, 1982-1998.  
41 *Global Biogeochemical Cycles*, 16(2), doi:10.1029/2001GB001550.
- 42 Hicke, J.A., G.P. Asner, J.T. Randerson, C. Tucker, S. Los, R. Birdsey, J.C. Jenkins, C. Field, and E.  
43 Holland, 2002b: Satellite-derived increases in net primary productivity across North America, 1982-  
44 1998. *Geophysical Research Letters*, 29(10), doi:10.1029/2001GL013578.
- 45 Hicke, J.A., G.P. Asner, E.S. Kasichke, N.H.F. French, J.T. Randerson, G.J. Collatz, B.J. Stocks, C.J.  
46 Tucker, S.O. Los, and C.B. Field, 2003: Postfire response of North American boreal forest net primary  
47 productivity analyzed with satellite observations. *Global Change Biology*, 9(8), 1145-1157.
- 48 Hild, L., A. Richter, V. Rozanov, and J.P. Burrows, 2002: Air mass factor calculations for GOME  
49 measurements of lightning-produced NO<sub>2</sub>. *Advances in Space Research*, 29, 1685-1690.
- 50 Hobbie, S.E., J.P. Schimel, S.E. Trumbore, and J.R. Randerson, 2000: Controls over carbon storage and  
51 turnover in high-latitude soils. *Global Change Biology*, 6, 196-210.
- 52 Hoerling, M., and A. Kumar, 2003: The perfect ocean for drought. *Science*, 299(5607), 691-694.
- 53 Holland, E.A., and M.A. Carroll, 2003: Atmospheric chemistry and the bio-atmospheric carbon and nitrogen  
54 cycles. In: *Interactions of the Major Biogeochemical Cycles, Global Change and Human Impacts*  
55 [Melillo, J.M., C.B. Field, and B. Moldan eds.]. SCOPE volume 61, Island Press, Washington, pp 273-  
56 294.

- 1 Holland, E.A., F.J. Dentener, B.H. Braswell, and J.M. Sulzman, 1999: Contemporary and pre-industrial  
2 reactive nitrogen budgets. *Biogeochemistry*, 46, 7-43.
- 3 Holland, E.A., B.H. Braswell, J. Sulzman, and J.F. Lamarque, 2005: Nitrogen deposition onto the United  
4 States and Western Europe: synthesis of observations and models. *Ecological Applications*, 15(1), 38-  
5 57.
- 6 Holzer, M., and G.J. Boer, 2001: Simulated changes in atmospheric transport climate. *Journal of Climate*,  
7 14, 4398-4420.
- 8 Hoppema, M., 2004: Weddell Sea turned from source to sink for atmospheric CO<sub>2</sub> between pre-industrial  
9 time and present. *Global and Planetary Change*, 40, 219-231.
- 10 Horowitz, L.W., S. Walters, D.L. Mauzerall, L.K. Emmons, P.J. Rasch, J. Philip, C. Granier, X. Tie, J.-F.  
11 Lamarque, M.G. Schultz, G.S. Tyndall, J.J. Orlando, and G.P. Brasseur, 2003: A global simulation of  
12 tropospheric ozone and related tracers: description and evaluation of MOZART, version 2. *Journal of*  
13 *Geophysical Research*, 108, 4784, doi:10.1029/2002JD002853.
- 14 Houghton, R.A., 2000: Interannual variability in the global carbon cycle. *Journal of Geophysical Research*,  
15 105(D15), 20121-20130, doi:10.1029/2000JD900041.
- 16 Houghton, R.A., 2003: Revised estimates of the annual net flux of carbon to the atmosphere from changes in  
17 land use and land management 1850-2000. *Tellus Series B*, 55(2), 378-390.
- 18 Houghton, R.A., 2003: Why are estimates of the terrestrial carbon balance so different? *Global Change*  
19 *Biology*, 9(4), 500-509.
- 20 Houghton, R.A., D.L. Skole, C.A. Nobre, J.L. Hackler, K.T. Lawrence, and W.H. Chomentowski, 2000:  
21 Annual fluxes of carbon from deforestation and regrowth in the Brazilian Amazon. *Nature*, 403, 301-  
22 304.
- 23 House, J.I., I.C. Prentice, and C. Le Quéré, 2002: Maximum impacts of future reforestation or deforestation  
24 on atmospheric CO<sub>2</sub>. *Global Change Biology*, 8(11), 1047-1052.
- 25 House, J.I., I.C. Prentice, N. Ramankutty, R.A. Houghton, and M. Heimann, 2003: Reconciling apparent  
26 inconsistencies in estimates of terrestrial CO<sub>2</sub> sources and sinks. *Tellus Series B*, 55(2), 345-363.
- 27 Houweling, S., F. Dentener, and J. Lelieveld, 1998: The impact of nonmethane hydrocarbon compounds on  
28 tropospheric photochemistry. *Journal of Geophysical Research*, 103, 10673-10696,  
29 doi:10.1029/97JD03582.
- 30 Hu, H., 1996: Water vapour and temperature lapse rate feedbacks in the mid-latitude seasonal cycle.  
31 *Geophysical Research Letters*, 23, 1761-1764.
- 32 Huang, Y., R. Dickinson, and W. Chameides, 2005: Impact of aerosol indirect effect on climate over East  
33 Asia. *Proceeding of the National Academy of Sciences USA*, (submitted).
- 34 Hudman, R.C., D.J. Jacob, O.R. Cooper, M.J. Evans, C.L. Heald, R.J. Park, F. Fehsenfeld, F. Flocke, J.  
35 Holloway, G. Hübler, K. Kita, M. Koike, Y. Kondo, A. Neuman, J. Nowak, S. Oltmans, D. Parrish,  
36 J.M. Roberts, T. Ryerson, 2004: Ozone production in transpacific Asian pollution plumes and  
37 implications for ozone air quality in California. *Journal of Geophysical Research*, 109, D23S10,  
38 doi:10.1029/2004JD004974.
- 39 Huesemann, M.H., A.D. Skillman, and E.A. Crecelius, 2002: The inhibition of marine nitrification by ocean  
40 disposal of carbon dioxide. *Marine Pollution Bulletin*, 44(2), 142-148.
- 41 Hughes, T.P., A.H. Baird, D.R. Bellwood, M. Card, S.R. Connolly, C. Folke, R. Grosberg, O. Hoegh-  
42 Guldborg, J.B.C. Jackson, J. Kleypas, J.M. Lough, P. Marshall, M. Nyström, S.R. Palumbi, J.M.  
43 Pandolfi, B. Rosen, and J. Roughgarden, 2003: Climate change, human impacts, and the resilience of  
44 coral reefs. *Science*, 301, 929-933.
- 45 Hulme, M., T.J. Osborn, and T.C. Johns, 1998: Precipitation sensitivity to global warming: comparison of  
46 observations with HadCM2 simulations. *Geophysical Research Letters*, 25, 3379-3382.
- 47 Humborg, C., 2000: Silicon retention in river basins: far-reaching effects on biogeochemistry and aquatic  
48 food webs in coastal marine environments. *Ambio*, 29(1), 45-50.
- 49 Hungate, B., J.S. Dukes, M.R. Shaw, Y. Luo, and C.B. Field, 2003: Nitrogen and climate change. *Science*,  
50 302(5650), 1512-1513.
- 51 Huntingford, C., P. Harris, N. Gedney, P. Cox, R. Betts, J. Marengo, and J. Gash, 2004: Using a GCM  
52 analogue model to investigate the potential for Amazonian forest dieback. *Theoretical and Applied*  
53 *Climatology*, 78(1-3), 177-185.
- 54 Hurtt, G.C., S.W. Pacala, P.R. Moorcroft, J. Caspersen, E. Shevliakova, R.A. Houghton, and B. Moore III,  
55 2002: Projecting the future of the U.S. carbon sink. *Proceedings of the National Academy of Sciences*  
56 *USA*, 99(3), 1389-1394.

- 1 Hutchins, D.A., and K.W. Bruland, 2000: Iron-limited diatom growth and Si:N uptake ratios in a coastal  
2 upwelling regime. *Nature*, 393, 561-564.
- 3 Hutyra, L., J.W. Munger, C.A. Nobre, S.R. Saleska, S.A. Vieira, S.C. Wofsy, 2005: Climatic variance and  
4 vulnerability to drought in Amazonia. *Geophysical Research Letters*, (submitted).
- 5 IPCC, 2000: *Emissions scenarios: special report of the Intergovernmental Panel on Climate Change*  
6 (*IPCC*). [Nakicenovic, N., and R. Swart (eds.)]. Cambridge University Press, Cambridge, 570 pp.
- 7 Irannejad, P., A. Henderson-Sellers, and S. Sharmeen, 2003: Importance of land-surface parameterisation for  
8 latent heat simulation in global atmospheric models. *Geophysical Research Letters*, 30(17), 1904,  
9 doi:10.1029/2003GL018044.
- 10 Ittekkot, V., 1993: The abiotically driven biological pump in the ocean and short-term fluctuations in  
11 atmospheric CO<sub>2</sub> contents. *Global and Planetary Change*, 8(1-2), 17-25.
- 12 Jacob, D.J., 2000: Heterogeneous chemistry and tropospheric ozone. *Atmospheric Environment*, 34, 2131-  
13 2159.
- 14 Jacob, D.J., J.A. Logan, G.M. Gardner, R.M. Yevich, C.M. Spivakovsky, S.C. Wofsy, S. Sillman, and M.J.  
15 Prather, 1993: Factors regulating ozone over the United States and its export to the global atmosphere.  
16 *Journal of Geophysical Research*, 98, 14817-14826.
- 17 Jaeglé, L., D.J. Jacob, W.H. Brune, and P.O. Wennberg, 2001: Chemistry of HOx radicals in the upper  
18 troposphere. *Atmospheric Environment*, 35, 469-489.
- 19 Jaenecke, R., 2005: Abundance of cellular material and proteins in the atmosphere. *Science*, 308(5718),  
20 doi:10.1126/science.1106335.
- 21 Jaffe, D., I. Bertsch, L. Jaegle, P. Novelli, J.S. Reid, H. Tanimoto, R. Vingarzan, and D.L. Westphal, 2004:  
22 Long-range transport of Siberian biomass burning emissions and impact on surface ozone in western  
23 North America. *Geophysical Research Letters*, 31, L16106, doi:10.1029/2004GL020093.
- 24 Jassby, A.D., and T. Platt, 1976: Mathematical formulation of the relationship between photosynthesis and  
25 light for phytoplankton. *Limnology and Oceanography*, 21, 540-547.
- 26 Jenny, H., 1941: Factors of soil formation: a system of quantitative pedology. McGraw-Hill, New York.
- 27 Jin, M., and R.E. Dickinson, 2002: New observational evidence for global warming from satellite data set.  
28 *Geophysical Research Letters*, 29(10), doi:10.1029/2001GLO013833.
- 29 Jin, M.L., J.M. Shepherd, and M.D. King, 2005: Urban aerosols and their variations with clouds and rainfall:  
30 a case study for New York and Houston. *Journal of Geophysical Research*, 110,  
31 doi:10.1029/2004JD005081.
- 32 Jin, Y., C. Schaaf, F. Gao, X. Li, A. Strahler, X. Zeng, R. Dickinson, 2002: How does snow impact the  
33 albedo of vegetated land surfaces as analyzed with MODIS data? *Geophysical Research Letters*, 29,  
34 doi:10.1029/2001GLO14132.
- 35 Johns, T., C. Durman, H. Banks, M. Roberts, A. McLaren, J. Ridley, C. Senior, K. Williams, A. Jones, A.  
36 Keen, G. Rickard, S. Cusack, M. Joshi, M. Ringer, B. Dong, H. Spencer, R. Hill, J. Gregory, A.  
37 Paradaens, J. Lowe, A. Bodas-Salcedo, S. Stark, and Y. Searl, 2004: *HadGEM1 – Model description and*  
38 *analysis of preliminary experiments for the IPCC Fourth Assessment Report*, 55. Hadley Centre, 75 pp.
- 39 Johnson, C.E., W.J. Collins, D.S. Stevenson, and R.G. Derwent, 1999: Relative roles of climate and  
40 emissions changes on future tropospheric oxidant concentrations. *Journal of Geophysical Research*,  
41 104(D15), 18631-18645.
- 42 Johnson, C.E., D.S. Stevenson, W.J. Collins, and R.G. Derwent, 2001: Role of climate feedback on methane  
43 and ozone studied with a coupled ocean-atmosphere-chemistry model. *Geophysical Research Letters*,  
44 28(9), 1723-1726.
- 45 Johnson, C.E., D.S. Stevenson, W.J. Collins, and R.G. Derwent, 2002: Interannual variability in methane  
46 growth rate simulated with a coupled ocean-atmosphere-chemistry model. *Geophysical Research*  
47 *Letters*, 29(19), 1903, doi:10.1029/2002GL015269.
- 48 Jones, A., D.L. Roberts, and M.J. Woodage, 2001: Indirect sulphate aerosol forcing in a climate model with  
49 an interactive sulphur cycle. *Journal of Geophysical Research*, 106, 20293-30310.
- 50 Jones, C.D., and P.M. Cox, 2001: Modelling the volcanic signal in the atmospheric CO<sub>2</sub> record. *Global*  
51 *Biogeochemical Cycles*, 15(2), 453-466.
- 52 Jones, C. D., and P. M. Cox, 2005: On the significance of atmospheric CO<sub>2</sub> growth rate anomalies in 2002–  
53 2003, *Geophysical Research Letters*, 32, L14816, doi:10.1029/2005GL023027.
- 54 Jones, C.D., M. Collins, P.M. Cox, and S.A. Spall, 2001: The carbon cycle response to ENSO: a coupled  
55 climate-carbon cycle model study. *Journal of Climate*, 14, 4113-4129.

- 1 Jones, C.D., P.M. Cox, R.L.H. Essery, D.L. Roberts, and M. Woodage, 2003a: Strong carbon cycle  
2 feedbacks in a climate model with interactive CO<sub>2</sub> and sulphate aerosols. *Geophysical Research Letters*,  
3 30(9), 1479, doi:10.1029/2003GL016867.
- 4 Jones, C.D., P.M. Cox, and C. Huntingford, 2003b: Uncertainty in climate-carbon cycle projections  
5 associated with the sensitivity of soil respiration to temperature. *Tellus B*, 55(2), 692-700.
- 6 Jones, C., N. Mahowald, and C. Luo, 2004: Observational evidence of African desert dust intensification of  
7 easterly waves. *Geophysical Research Letters*, 31, doi:10.1029/2004GL020107.
- 8 Jones, C.D., C. McConnell, K. Coleman, P. Cox, P. Falloon, D. Jenkinson, and D. Powlson, 2005a: Global  
9 climate change and soil carbon stocks: predictions from two contrasting models for the turnover of  
10 organic carbon in soil. *Global Change Biology*, 11(1), 154-166.
- 11 Jones, C.D., C.L. McConnell, K. Coleman, P.M. Cox, P. Falloon, D.S. Jenkinson, and D.S. Powlson, 2005b:  
12 The sensitivity of projected soil carbon storage under climate change to the representation of soil carbon  
13 dynamics. *Global Change Biology*, 11, 154-166
- 14 Joos, F., I. C. Prentice, S. Sitch, R. Meyer, G. Hooss, G.-K. Plattner, S. Gerber, and K. Hasselmann, 2001:  
15 Global warming feedbacks on terrestrial carbon uptake under the Intergovernmental Panel on Climate  
16 Change (IPCC) emission scenarios. *Global Biogeochemical Cycles*, 15, 891-907.
- 17 Joshi, M.M., K. Shine, M. Ponater, N. Stuber, R. Sausen, and L. Li, 2003: A comparison of climate response  
18 to different radiative forcings in three general circulation models: Towards an improved metric of  
19 climate change. *Climate Dynamics*, 20, 843-854.
- 20 Kalnay, E., and M. Cai, 2003: Impact of urbanization and land-use change on climate. *Nature*, 423, 528-531.
- 21 Kalnay, E., M. Cai, and H. Li, 2004: Estimation of the impact of land-use changes and urbanization on  
22 climate trends east of the Rockies. *Journal of Geophysical Research* (in press).
- 23 Kaminski, T., M. Heimann, and R. Giering, 1999: A coarse grid three-dimensional global inverse model of  
24 the atmospheric transport, 2. Inversion of the transport of CO<sub>2</sub> in the 1980s. *Journal of Geophysical  
25 Research*, 104(D15), 18555-18581.
- 26 Kanakidou, M., J. Seinfeld, S. Pandis, I. Barnes, F. Dentener, M. Facchini, R.v. Dingenen, B. Ervens, A.  
27 Nenes, C. Nielsen, E. Swietlicki, J.P. Putaud, Y. Balkanski, S. Fuzzi, J. Horth, G.K. Moortgat, R.  
28 Winterhalter, C.E.L. Myhre, K. Tsigaridis, E. Vignati, E.G. Stephanou, and J. Wilson, 2004: Organic  
29 aerosol and global climate modelling: a review. *Atmospheric Chemistry & Physics Discussions*, 4,  
30 5855-6024.
- 31 Kaplan, J.O., I.C. Prentice, W. Knorr, and P.J. Valdez, 2002: Modelling the dynamics of terrestrial carbon  
32 storage since the Last Glacial Maximum. *Geophysical Research Letters*, 29(22), 2074,  
33 doi:10.1029/2002GL015230.
- 34 Kärcher, B., and U. Lohmann, 2002: A parameterization of cirrus cloud formation: homogeneous freezing of  
35 supercooled aerosols. *Journal of Geophysical Research*, 107, doi:10.1029/2001JD000470.
- 36 Kärcher, B., and U. Lohmann, 2003: A parameterization of cirrus cloud formation: heterogeneous freezing.  
37 *Journal of Geophysical Research*, 108, doi:10.1029/2002JD003220.
- 38 Kärcher, B., and T. Koop, 2004: The role of organic aerosols in homogeneous ice formation. *Atmospheric  
39 Chemistry & Physics Discussions*, 4, 6719-6745.
- 40 Kärcher, B., and J. Ström, 2003: The roles of dynamical variability and aerosols in cirrus cloud formation.  
41 *Atmospheric Chemistry & Physics*, 3, 823-838.
- 42 Karlsdottir, S., and I.S.A. Isaksen, 2000: Changing methane lifetime: Possible cause for reduced growth.  
43 *Geophysical Research Letters*, 27(1), 93-96.
- 44 Kasibhatla, P., A. Arellano, J.A. Logan, P.I. Palmer, and P. Novelli, 2002: Top-down estimate of a large  
45 source of atmospheric carbon monoxide associated with fuel combustion in Asia. *Geophysical Research  
46 Letters*, 29(19), 1900, doi:10.1029/2002GL015581.
- 47 Kasischke, E.S., and L.P. Bruhwiler, 2002: Emissions of carbon dioxide, carbon monoxide, and methane  
48 from boreal forest fires in 1998. *Journal of Geophysical Research*, 107, 8146,  
49 doi:10.1029/2001JD000461 [printed 108 (D1), 2003].
- 50 Kasischke, E.S., and J.E. Penner, 2004: Improving global estimates of atmospheric emissions from biomass  
51 burning. *Journal of Geophysical Research-Atmospheres*, 109, D14S01, doi:10.1029/2004JD004972.
- 52 Kasischke, E.S., N.L. Christensen, and B.J. Stocks, 1995: Fire, global warming and the carbon balance of  
53 boreal forests. *Ecological Applications*, 5(2), 437-451.
- 54 Kasischke, E.S., E.J. Hyer, P.C. Novelli, L.P. Bruhwiler, N.H.F. French, A.I. Sukhinin, J.H. Hewson, and  
55 B.J. Stocks, 2005: Influences of boreal fire emissions on Northern Hemisphere atmospheric carbon and  
56 carbon monoxide. *Global Biogeochemical Cycles*, 19(1), GB1012, doi:10.1029/2004GB002300.

- 1 Katul, G.G., L. Mahrt, D. Poggi, and C. Sanz, 2004: One- and two-equation models for canopy turbulence.  
2 *Boundary-Layer Meteorology*, 113, 81-109.
- 3 Kawamiya, M., C. Yoshikawa, H. Sato, K. Sudo, S. Watanabe, and T. Matsuno, 2005: Development of an  
4 integrated earth system model on the Earth Simulator. *Journal of Earth Simulator*, (in prep.)
- 5 Keeling, C.D., and S.C. Piper, 2001: *Exchanges of atmospheric CO<sub>2</sub> and <sup>13</sup>CO<sub>2</sub> with the terrestrial biosphere  
6 and oceans from 1978 to 2000. IV. Critical overview*. SIO Reference Series 01-09, pp. 1-23.
- 7 Keeling, C.D., and T.P. Whorf, 2004: Atmospheric CO<sub>2</sub> records from sites in the SIO air sampling network.  
8 In: *Trends Online: A Compendium of Data on Global Change*. Carbon Dioxide Information Analysis  
9 Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tenn., U.S.A.
- 10 Keeling, R.F., 2005: Comment on "The ocean sink for anthropogenic CO<sub>2</sub>". *Science*, 308,  
11 doi:10.1126/science.1109620.
- 12 Keller, M., and W.A. Reiner, 1994: Soil-atmosphere exchange of nitrous oxide, nitric oxide, and methane  
13 under secondary succession of pasture to forest in the Atlantic lowlands of Costa Rica. *Global  
14 Biogeochemical Cycles*, 8, 399-409.
- 15 Kesselmeier, J., P. Ciccioli, U. Kuhn, P. Stefani, T. Biesenthal, S. Rottenberger, A. Wolf, M. Vitullo, R.  
16 Valentini, A. Nobre, P. Kabat, and M.O. Andreae, 2002: Volatile organic compound emissions in  
17 relation to plant carbon fixation and the terrestrial carbon budget. *Global Biogeochemical Cycles*,  
18 16(4),1126, doi:10.1029/2001GB001813.
- 19 Kettle, A., and M. Andreae, 2000: Flux of the dimethylsulfide from the oceans: A comparison of updated  
20 data sets and flux models. *Journal of Geophysical Research*, 105, 26793-26808.
- 21 Khain, A.P., D. Rosenfeld, and A. Pokrovsky, 2001: Simulating convective clouds with sustained  
22 supercooled liquid water down to -37.5°C using a spectral microphysics model. *Geophysical Research  
23 Letters*, 28, 3887-3890.
- 24 Khain, A., A. Pokrovsky, M. Pinsky, A. Seifert, and V. Phillips, 2004: Simulation of effects of atmospheric  
25 aerosols on deep turbulent convective using a spectral microphysics mixed-phase cumulus cloud model.  
26 1. Model description and possible applications. *Journal of Aerosol Science*, 61, 2963-2982.
- 27 Khain, A., D. Rosenfeld, and A. Pokrovsky, 2005: Aerosol impact on the dynamics and microphysics of  
28 convective clouds. *Quarterly Journal of the Royal Meteorological Society*, (in press).
- 29 Khalil, M.A.K., and R.A. Rasmussen, 1993: Decreasing trend of methane: unpredictability of future  
30 concentrations. *Chemosphere*, 26, 803-814.
- 31 Khalil, M.A.K., M.J. Shearer, and R.A. Rasmussen, 2000: *Atmospheric methane: its role in the global  
32 environment*. [Khalil, M.A.K. (ed.)]. Springer, New York, pp. 86-97.
- 33 Kinne, S., M. Schulz, C. Textor, S. Guibert, Y. Balkanski, S. Bauer, T. Berntsen, T. Berglen, O. Boucher, M.  
34 Chin, W. Collins, F. Dentener, T. Diehl, R. Easter, J. Feichter, D. Fillmore, S. Ghan, P. Ginoux, S.  
35 Gong, A. Grini, J. Hendricks, M. Herzog, L. Horowitz, I. Isaksen, T. Iversen, A. Jones, S. Kloster, D.  
36 Koch, M. Krool, A. Lauer, J.F. Lamarque, G. Lesins, X. Liu, U. Lohmann, V. Montanaro, G. Myhre, J.  
37 Penner, G. Pitari, S. Reddy, D. Roberts, O. Seland, P. Stier, T. Takemura, and X. Tie, 2005: An  
38 AeroCom initial assessment – optical properties in aerosol component modules of global models.  
39 *Atmospheric Chemistry & Physics Discussions*, (submitted).
- 40 Kiorboe, T., J.L.S. Hansen, A.L. Alldredge, G.A. Jackson, U. Passow, H.G. Dam, D.T. Drapeau, A. Waite,  
41 and C.M. Garcia, 1996: Sedimentation of phytoplankton during a diatom bloom: rates and mechanisms.  
42 *Journal of Marine Research*, 54(6), 1123-1148.
- 43 Klaas, C., and D.E. Archer, 2002: Association of sinking organic matter with various types of mineral ballast  
44 in the deep sea: Implications for the rain ratio. *Global Biogeochemical Cycles*, 16(4), 1116,  
45 doi:10.1029/2001GB001765.
- 46 Kleypas, J.A., R.W. Buddemeier, D. Archer, J.-P. Gattuso, C. Langdon, and B.N. Opdyke, 1999:  
47 Geochemical consequences of increased atmospheric carbon dioxide on coral reefs. *Science*, 284, 118-  
48 120.
- 49 Knief, C., S. Vanitchung, N.V. Harvey, R. Conrad, P.F. Dunfield, and A. Chidthaisong, 2005: Diversity of  
50 methanotrophic bacteria in tropical upland soils under different land uses. *Applied and Environmental  
51 Microbiology*, 71, 3862-3831.
- 52 Knorr, W., I.C. Prentice, J.I. House, and E.A. Holland, 2005: Long-term sensitivity of soil carbon turnover to  
53 warming. *Nature*, 433, 298-301.
- 54 Knowlton, K., J.E. Rosenthal, C. Hogrefe, B. Lynn, S. Gaffin, R. Goldberg, C. Rosenzweig, K. Civerolo,  
55 J.Y. Ku, and P.L. Kinney, 2004: Assessing ozone-related health impacts under a changing climate.  
56 *Environmental Health Perspectives*, 112, 1557-1563.

- 1 Koerner, C., 2004: Through enhanced tree dynamics carbon dioxide enrichment may cause tropical forests to  
2 lose carbon. *Philosophical Transactions of the Royal Society of London, Series B*, 359, 493-498.
- 3 Koster, R.D., and M.J. Suarez, 2001: Soil moisture memory in climate models. *Journal of*  
4 *Hydrometeorology*, 2(6), 558-570.
- 5 Koster, R.D., and M.J. Suarez, 2004: Suggestions in the observational record of land-atmosphere feedback  
6 operating at seasonal time scales. *Journal of Hydrometeorology*, 5(3), doi: 10.1175/1525.
- 7 Koster, R.D., M. Suarez, A. Ducharne, M. Stieglitz, and P. Kumar, 2000: A catchment-based approach to  
8 modeling land surface processes in a general circulation model 1. Model structure. *Journal of*  
9 *Geophysical Research*, 105, 24809-24822.
- 10 Koster, R.D., P.A. Dirmeyer, A.N. Hahmann, R. Ijpehaar, L. Tyahla, P. Cox, and M.J. Suarez, 2002:  
11 Comparing the degree of land - atmosphere interaction in four atmospheric general circulation models.  
12 *Journal of Hydrometeorology*, 3(3), 363-375.
- 13 Koster, R.D., M.J. Suarez, R.W. Higgins, and H.M. Van den Dool, 2003: Observational evidence that soil  
14 moisture variations affect precipitation. *Geophysical Research Letters*, 30(5), 1241.  
15 doi:10.1029/2002GL016571,
- 16 Krakauer, N.Y., and J.T. Randerson, 2003. Do volcanic eruptions enhance or diminish net primary  
17 production? Evidence from tree rings. *Global Biogeochemical Cycles*, 17(4), 1118,  
18 doi:10.1029/2003GB002076.
- 19 Kristjánsson, J.E., 2002: Studies of the aerosol indirect effect from sulphate and black carbon aerosols.  
20 *Journal of Geophysical Research*, 107, doi:10.1029/2001JD000887.
- 21 Krol, M., and J. Lelieveld, 2003: Can the variability in tropospheric OH be deduced from measurements of  
22 1,1,1-trichloroethane (methyl chloroform)? *Journal of Geophysical Research*, 108(D3), 4125,  
23 doi:10.1029/2001JD002040.
- 24 Krüger, O., and H. Graßl, 2002: The indirect aerosol effect over Europe. *Geophysical Research Letters*, 29,  
25 doi:10.1029/2001GL014081.
- 26 Kuma, K., J. Nishioka, and K. Matsunaga, 1996: Controls on iron (III) hydroxide solubility in seawater. The  
27 influence of pH and natural organic chelators. *Limnology and Oceanography*, 41, 396-407.
- 28 Kurz, W.A., and M.J. Apps, 1999: A 70-year retrospective analysis of carbon fluxes in the Canadian forest  
29 sector. *Ecological Applications*, 9(2), 526-547.
- 30 Kurz, W.A., M.J. Apps, S.J. Beukema, and T. Lekstrum, 1995: 20th-century carbon budget of Canadian  
31 forests. *Tellus Series B*, 47(1-2), 170-177.
- 32 Kvenvolden, K.A., 1993: Gas hydrates: Geological perspective and global change. *Reviews of Geophysics*,  
33 31, 173-187.
- 34 Kvenvolden, K.A., 2000: Gas hydrates and humans. *Annals of the New York Academy of Sciences*, 912, 17-  
35 22.
- 36 Kvenvolden, K.A., 2002: Methane hydrate and the global organic carbon cycle. *Terra Nova*, 14(5), 302-306.
- 37 Labrador, L.J., R. von Kuhlmann, and M.G. Lawrence, 2004: Strong sensitivity of the global mean OH  
38 concentration and the tropospheric oxidizing efficiency to the source of NO<sub>x</sub> from lightning.  
39 *Geophysical Research Letters*, 31, L06102, doi:10.1029/2003GL019229.
- 40 Lamarque, J.-F., and P.G. Hess, 2004: Arctic Oscillation modulation of the northern hemisphere spring  
41 tropospheric ozone. *Geophysical Research Letters*, 31, L06127, doi:10.1029/2003GL019116.
- 42 Lamarque, J.-F., J.T. Kiehl, P.G. Hess, W.D. Collins, L.K. Emmons, P. Ginoux, C. Luo, and X.X. Tie, 2005:  
43 Coupled chemistry-climate response to changes in aerosol emissions: global impact on the hydrological  
44 cycle and the tropospheric burdens of OH, ozone and NO<sub>x</sub>. *Geophysical Research Letters*, 32, L16809,  
45 doi: 10.1029/2005GL023419.
- 46 Lamarque, J.-F., P. Hess, L. Emmons, L. Buja, W. Washington, and C. Granier, 2005: Tropospheric ozone  
47 evolution between 1890 and 1990. *Journal of Geophysical Research*, 110, D08304,  
48 doi:10.1029/2004JD005537.
- 49 Lance, S., A. Nenes, and T.A. Rissman, 2004: Chemical and dynamical effects on cloud droplet number:  
50 implications for estimates of the aerosol indirect effect. *Journal of Geophysical Research*, 109,  
51 doi:10.1029/2004JD004596.
- 52 Langenfelds, R.L., R.J. Francey, B.C. Pak, L.P. Steele, J. Lloyd, C.M. Trudinger, and C.E. Allison, 2002:  
53 Interannual growth rate variations of atmospheric CO<sub>2</sub> and its <sup>13</sup>C, H<sub>2</sub>, CH<sub>4</sub>, and CO between 1992 and  
54 1999 linked to biomass burning. *Global Biogeochemical Cycles*, 16(3), 1048,  
55 doi:10.1029/2001GB001466.
- 56 Lara, L.L., E. A. Holland, P. Artaxo, P. B. Camargo, and L. A. Martinelli, 2005: Linking nitrogen deposition  
57 patterns and land use change in tropical areas. *Biogeochemistry*, (in press).

- 1 Lassey, K.R., D.C. Lowe, and M.R. Manning, 2000: The trend in atmospheric methane <sup>13</sup>C and implications  
2 for isotopic constraints on the global methane budget. *Global Biogeochemical Cycles*, 14, 41-49.
- 3 Laurance, W.F., A.A. Oliveira, S.G. Laurance, R. Condit, H.E.M. Nascimento, A.C. Sanchez-Thorin, T.E.  
4 Lovejoy, A. Andrade, S. D'Angelo, J.E. Ribeiro, and C.W. Dick, 2004: Pervasive alteration of tree  
5 communities in undisturbed Amazonian forests. *Nature*, 428, 171-175.
- 6 Law, R.M., Y-H. Chen, K.R. Gurney, and TRANSCOM 3 modellers, 2003: TransCom 3 CO<sub>2</sub> inversion  
7 intercomparison: 2. Sensitivity of annual mean results to data choices. *Tellus B*, 55(2), 580-595.
- 8 Lawrence, D.M., and J.M. Slingo, 2004: An annual cycle of vegetation in a GCM. Part I: implementation  
9 and impact on evaporation. *Climate Dynamics*, 22, doi:10.1007/s0038200303669.
- 10 Lawrence, M.G., P. Jockel, and R. von Kuhlmann, 2001: What does the global mean OH concentration tell  
11 us? *Atmospheric Chemistry & Physics*, 1, 37-49.
- 12 Lawrence, M.G., R. von Kuhlmann, M. Salzmann, and P.J. Rasch, 2003: The balance of effects of deep  
13 convective mixing on tropospheric ozone. *Geophysical Research Letters*, 30(18), 1940,  
14 doi:10.1029/2003GL017644.
- 15 Leaitch, W.R., K. Hayden, W. Strapp, D. Toom-Sauntry, M. Wasey, U. Lohmann, P. Lehr, J. Marshall, W.  
16 Miller, N. Shantz, M. Mozurkewich, D. Waugh, T. Garrett, J. Jayne, D. Worsnop, and M. Wolde, 2005:  
17 Evidence for an indirect effect of the organic aerosol. *Journal of Geophysical Research*, 110,  
18 (submitted).
- 19 Leck, C., and K. Bigg, 2005: Biogenic particles over the central Arctic Ocean. *Tellus*, 57B, (in press).
- 20 Lee, K., R. Wanninkhof, T. Takahashi, S.C. Doney, and R.A. Feely, 1998: Low interannual variability in  
21 recent oceanic uptake of atmospheric carbon dioxide. *Nature*, 396, 155-159.
- 22 Lee, K., S.D. Choi, G.H. Park, R. Wanninkhof, T.H. Peng, R.M. Key, C.L. Sabine, R.A. Feely, J.L.  
23 Bullister, F.J. Millero, and A. Kozyr, 2003: An updated anthropogenic CO<sub>2</sub> inventory in the Atlantic  
24 Ocean. *Global Biogeochemical Cycles*, 17(4), 1116, doi:10.1029/2003GB002067.
- 25 Lefevre, N., A.J. Watson, A. Olsen, A.F. Rios, F.F. Pérez, and T. Johannessen, 2004: A decrease in the sink  
26 for atmospheric CO<sub>2</sub> in the North Atlantic. *Geophysical Research Letters*, 31(7), L07306,  
27 doi:10.1029/2003GL018957.
- 28 Lelieveld, J., and F.J. Dentener, 2000: What controls tropospheric ozone? *Journal of Geophysical Research*,  
29 105, 3531-3551.
- 30 Lelieveld, J., P.J. Crutzen, and F.J. Dentener, 1998: Changing concentration, lifetime and climate forcing of  
31 atmospheric methane. *Tellus Series B*, 50(2), 128-150.
- 32 Lelieveld, J., W. Peters, F.J. Dentener, and M.C. Krol, 2002a: Stability of tropospheric hydroxyl chemistry.  
33 *Journal of Geophysical Research-Atmospheres*, 107(D23), 4715, doi:10.1029/2002JD002272.
- 34 Lelieveld, J., H. Berresheim, S. Borrmann, P.J. Crutzen, F.J. Dentener, H. Fischer, J. Feichter, P.J. Flatau, J.  
35 Heland, R. Holzinger, R. Kormann, M.G. Lawrence, Z. Levin, K.M. Markowicz, N. Mihalopoulos, A.  
36 Minikin, V. Ramanathan, M. de Reus, G.J. Roelofs, H.A. Scheeren, J. Sciare, H. Schlager, M. Schultz,  
37 P. Siegmund, B. Steil, E.G. Stephanou, P. Stier, M. Traub, C. Warneke, J. Williams, and H. Ziereis,  
38 2002b: Global air pollution crossroads over the Mediterranean. *Science*, 298, 794-799.
- 39 Lelieveld, J., S. Fuzzi, C. Granier, N. Harris, O. Hov, M. De Maziere, and U. Schumann, 2005: *Atmospheric*  
40 *Change and Earth System Science*. EUR Report No. 21461, European Commission, Rue de la Loi 200,  
41 B-1049 Brussels, Belgium.
- 42 LeQuéré, C., J.C. Orr, P. Monfray, O. Aumont, and G. Madec, 2000: Interannual variability of the oceanic  
43 sink of CO<sub>2</sub> from 1979 to 1997. *Global Biogeochemical Cycles*, 14, 1247-1265.
- 44 LeQuéré, C., O. Aumont, L. Bopp, P. Bousquet, P. Ciais, R. Francey, M. Heimann, C.D. Keeling, R.F.  
45 Keeling, H. Khesghi, P. Peylin, S.C. Piper, I.C. Prentice, and P.J. Rayner, 2003: Two decades of ocean  
46 CO<sub>2</sub> sink and variability. *Tellus Series B*, 55(2), 649-656.
- 47 Leue, C., M. Wenig, T. Wagner, O. Klimm, U. Platt, and B. Jahne, 2001: Quantitative analysis of NO<sub>x</sub>  
48 emissions from GOME satellite image sequences. *Journal of Geophysical Research*, 106, 5493-5505.
- 49 Levis, S., and G.B. Bonan, 2004: Simulating springtime temperature patterns in the community atmosphere  
50 model coupled to the community land model using prognostic leaf area. *Journal of Climate*, 17, 4531-  
51 4540.
- 52 Levis, S., G.B. Bonan, and C. Bonfils, 2004: Soil feedback drives the mid-Holocene North African monsoon  
53 northward in fully coupled CCSM2 simulations with a dynamic vegetation model. *Climate Dynamics*,  
54 23, doi:10.1007/s00382-004-0477-y.
- 55 Lewis, E.R., and S.E. Schwartz, 2005: Sea salt aerosol production: Mechanisms, methods, measurements,  
56 and models: A critical review, Geophysical Monograph Series, 152, AGU,

- 1 Lewis, S. L., Y. Malhi, and O.L. Phillips, 2005: Fingerprinting the impacts of global change on tropical  
2 forests. *Philosophical Transactions of the Royal Society of London, Series B*, 359,  
3 doi:10.1098/rstb.2003.1432.
- 4 Li, C., J. Qiu, S. Frolking, X. Xiao, W. Salas, B. Moore III, S. Boles, Y. Huang, and R. Sass, 2002: Reduced  
5 methane emissions from large-scale changes in water management of China's rice paddies during 1980-  
6 2000. *Geophysical Research Letters*, 29(3), doi: 10.1029/2002GL015370.
- 7 Li, Q.B., D.J. Jacob, I. Bey, P.I. Palmer, B.N. Duncan, B.D. Field, R.V. Martin, A.M. Fiore, R.M. Yantosca,  
8 D.D. Parrish, P.G. Simmonds, and S.J. Oltmans, 2002: Transatlantic transport of pollution and its  
9 effects on surface ozone in Europe and North America. *Journal of Geophysical Research*, 107(D13),  
10 doi:10.1029/2001JD001422.
- 11 Li, Q., D.J. Jacob, R. Park, Y. Wang, C.L. Heald, R. Hudman, R.M. Yantosca, R.V. Martin, and M. Evans,  
12 2005: North American pollution outflow and the trapping of convectively lifted pollution by upper-level  
13 anticyclone. *Journal of Geophysical Research*, 110, D10301, doi:10.1029/2004JD005039.
- 14 Liang X., Z. Xie, and M. Huang, 2003: A new parameterization for surface and groundwater interactions and  
15 its impact on water budgets with the variable infiltration capacity (VIC) land surface model. *Journal of*  
16 *Geophysical Research*, 108, 8613, doi:19.1029/2002JD003090.
- 17 Liepert, B.G., 2002: Observed reductions of surface solar radiation at sites in the United States and  
18 worldwide from 1961 to 1990. *Geophysical Research Letters*, 29, doi:10.1029/2002GL014910.
- 19 Liepert, B.G., and I. Tegen, 2002: Multidecadal solar radiation trends in the United States and Germany and  
20 direct tropospheric aerosol forcing. *Journal of Geophysical Research*, 107, doi:10.1029/2001JD000760.
- 21 Liepert, B.G., J. Feichter, U. Lohmann, and E. Roeckner, 2004: Can aerosols spin down the water cycle in a  
22 warmer and moister world. *Geophysical Research Letters*, 31, L06207, doi:10.1029/2003GL019060.
- 23 Lin, C.-Y.C., D.J. Jacob, and A.M. Fiore, 2001: Trends in exceedances of the ozone air quality standard in  
24 the continental United States, 1980-1998. *Atmospheric Environment*, 35, 3217-3228.
- 25 Lintner, B.R., 2002: Characterizing global CO<sub>2</sub> interannual variability with empirical orthogonal  
26 function/principal component (EOF/PC) analysis. *Geophysical Research Letters*, 29(19), 1921,  
27 doi:10.1029/2001GL014419.
- 28 Liousse, C., J.E. Penner, C. Chuang, J.J. Walton, H. Eddleman, and H. Cachier, 1996: A global three-  
29 dimensional model study of carbonaceous aerosols. *Journal of Geophysical Research*, 101, 19411-  
30 19432.
- 31 Liston, G.E., 2004: Representing subgrid snow cover heterogeneities in regional and global models. *Journal*  
32 *of Climate*, 17(6), 1381-1397.
- 33 Liu, H., D.J. Jacob, J.E. Dibb, A.M. Fiore, and R.M. Yantosca, 2004: Constraints on the sources of  
34 tropospheric ozone from <sup>210</sup>Pb-<sup>7</sup>Be-O<sub>3</sub> correlations. *Journal of Geophysical Research*, 109, D07306,  
35 doi:10.1029/2003JD003988.
- 36 Liu, H.P., J.T. Randerson, J. Lindfors, and F.S. Chapin, 2005: Changes in the surface energy budget after  
37 fire in boreal ecosystems of interior Alaska: An annual perspective. *Journal of Geophysical Research-*  
38 *Atmospheres*, 110, D13101, doi:10.1029/2004JD005158.
- 39 Liu, Q., and R.E. Dickinson, 2003: Use of a two-mode soil pore size distribution to estimate soil water transport in a  
40 land surface model. *Geophysical Research Letters*, 30(6), 1331, doi:10.1029/2002GLO16562.
- 41 Liu, X., and F.J. Millero, 2002: The solubility of iron in seawater. *Marine Chemistry*, 77, 43-54.
- 42 Liu, Y., and P.H. Daum, 2002: Indirect warming effect from dispersion forcing. *Nature*, 419, 580-581.
- 43 Liu, Y., P.H. Daum, and R. McGraw, 2004: An analytical expression for predicting the critical radius in the  
44 autoconversion parameterization. *Geophysical Research Letters*, 31, doi:10.1029/2003GL019117.
- 45 Logan, J.A., 1999: An analysis of ozonesonde data for the troposphere: recommendations for testing 3-D  
46 models, and development of a gridded climatology for tropospheric ozone. *Journal of Geophysical*  
47 *Research*, 104, 16115-16149.
- 48 Loh, A.N., J.E. Bauer, and E.R.M. Druffel, 2004: Variable ageing and storage of dissolved organic  
49 components in the open ocean. *Nature*, 430, 877-881.
- 50 Lohmann, U., 2002: A glaciation indirect aerosol effect caused by soot aerosols. *Geophysical Research*  
51 *Letters*, 29, doi:10.1029/2001GL014357.
- 52 Lohmann, U., 2004: Can anthropogenic aerosols decrease the snowfall rate? *Journal of Atmospheric Science*,  
53 61, 2457-2468.
- 54 Lohmann, U., and K. Diehl, 2005: Sensitivity studies of the importance of dust ice nuclei for the indirect  
55 aerosol effect on stratiform mixed-phase clouds. *Journal of Atmospheric Science*, 62, (submitted).
- 56 Lohmann, U., and J. Feichter, 1997: Impact of sulphate aerosols on albedo and lifetime of clouds: a  
57 sensitivity study with the ECHAM GCM. *Journal of Geophysical Research*, 102, 13685-13700.

- 1 Lohmann, U., and J. Feichter, 2001: Can the direct and semi-direct aerosol effect compete with the indirect  
2 effect on a global scale? *Geophysical Research Letters*, 28(1), 159-161, doi:10.1029/2000GL012051.
- 3 Lohmann, U., and J. Feichter, 2005: Global indirect aerosol effects: a review. *Atmospheric Chemistry &*  
4 *Physics*, 5, 715-737.
- 5 Lohmann, U., and B. Kärcher, 2002: First interactive simulations of cirrus clouds formed by homogeneous  
6 freezing in the ECHAM GCM. *Journal of Geophysical Research*, 107, doi:10.1029/2001JD000767.
- 7 Lohmann, U., and C. Leck, 2005: Importance of submicron surface active organic aerosols for pristine  
8 Arctic clouds. *Tellus*, 57B, 261-268.
- 9 Lohmann, U., and G. Lesins, 2002: Stronger constraints on the anthropogenic indirect aerosol effect.  
10 *Science*, 298, 1012-1016.
- 11 Lohmann, U., J. Feichter, J.E. Penner, and W.R. Leaitch, 2000: Indirect effect of sulphate and carbonaceous  
12 aerosols: a mechanistic treatment. *Journal of Geophysical Research*, 105, 12193-12206.
- 13 Lotsch, A., M.A. Friedl, B.T. Anderson, and C.J. Tucker, 2003: Coupled vegetation-precipitation variability  
14 observed from satellite and climate records. *Geophysical Research Letters*, 30(14),  
15 doi:10.1029/2003GL017506.
- 16 Lotsch, A., M.A. Friedl, B.T. Anderson, and C.J. Tucker, 2005: Response of terrestrial ecosystems to recent  
17 Northern Hemispheric drought. *Geophysical Research Letters*, 32(6), L06705, doi:  
18 10.1029/2004GL0220430.
- 19 Lovins, A.B., 2003: Hydrogen primer. *RMI Solutions Newsletter*, 19(2), 1-4, 36-39.
- 20 Lucht, W., L.C. Prentice, R.B. Myneni, S. Sitch, P. Friedlingstein, W. Cramer, P. Bousquet, W. Buermann,  
21 and B. Smith, 2002: Climatic control of the high-latitude vegetation greening trend and Pinatubo effect.  
22 *Science*, 296(5573), 1687-1689.
- 23 Luo, Y., S. Wan, D. Hui, and L. Wallace, 2001: Acclimatization of soil respiration to warming in a tall grass  
24 prairie. *Nature*, 413, 622-625.
- 25 Luo, Y., B. Su, W.S. Currie, J.S. Dukes, A. Finzi, U. Hartwig, B. Hungate, R.E. McMurtrie, R. Oren, W.J.  
26 Parton, D.E. Pataki, M.R. Shaw, D.R. Zak, and C.B. Field, 2004: Progressive nitrogen limitation of  
27 ecosystem responses to rising atmospheric carbon dioxide. *Bioscience*, 54(8), 731-739.
- 28 Mahowald, N.M., and L.M. Kiehl, 2003: Mineral aerosol and cloud interactions. *Geophysical Research*  
29 *Letters*, 30, doi:10.1029/2002GL016762.
- 30 Mahowald, N.M., and C. Luo, 2003: A less dusty future? *Geophysical Research Letters*, 30(7), 1903,  
31 doi:10.1029/2003GL017880.
- 32 Malhi, Y., and J. Grace, 2000: Tropical forests and atmospheric carbon dioxide. *Trends In Ecology &*  
33 *Evolution*, 15(8), 332-337.
- 34 Malhi, Y., and O.L. Phillips, 2004: Tropical forests and global atmospheric change: a synthesis.  
35 *Philosophical Transactions of the Royal Society B*, 359, doi:10.1098/rstb.2003.1449.
- 36 Malhi, Y., and J. Wright, 2004: Spatial patterns and recent trends in the climate of tropical rainforest regions.  
37 *Philosophical Transactions of the Royal Society B*, 359, doi:10.1098/ rstb.2003.1433.
- 38 Malhi, Y., T.R. Baker, O.L. Phillips, S. Almeida, E. Alvarez, L. Arroyo, J. Chave, C.I. Czimczik, A. Di  
39 Fiore, N. Higuchi, T.J. Killeen, S.G. Laurance, W.F. Laurance, S.L. Lewis, L.M.M. Montoya, A.  
40 Monteagudo, D.A. Neill, P.N. Vargas, S. Patino, N.C.A. Pitman, C.A. Quesada, R. Salomao, J.N.M.  
41 Silva, A.T. Lezama, R.V. Martinez, J. Terborgh, B. Vinceti, and J. Lloyd, 2004: The above-ground  
42 coarse wood productivity of 104 Neotropical forest plots. *Global Change Biology*, 10(5), 563-591.
- 43 Manning, A.C., 2001: *Temporal variability of atmospheric oxygen from both continuous measurements and*  
44 *a flask sampling network: Tools for studying the global carbon cycle*. Ph.D. Thesis, 233 pp., University  
45 of California, San Diego, La Jolla, Calif.
- 46 Manning, A.C., and R.F. Keeling, 2005: Global oceanic and land biotic carbon sinks from the Scripps  
47 atmospheric oxygen flask sampling network, (submitted).
- 48 Manning, M.R., D.C. Lowe, R.C. Moss, G.E. Bodeker, and W. Allan, 2005: Short term variations in the  
49 oxidizing power of the atmosphere. *Nature*, 436, 1001-1004.
- 50 Marani, M., E. Eltahir, and A. Rinaldo, 2001: Geomorphic controls on regional base flow. *Water Resources*  
51 *Research*, 37, 2619-2630.
- 52 Marengo, J., 2004: Interdecadal and long term-variations of rainfall in the Amazon basin. *Theoretical and*  
53 *Applied Climatology*, 78, 76-96.
- 54 Marengo, J., and C.A. Nobre, 2001: The hydroclimatological framework in Amazonia. In: *Biogeochemistry*  
55 *of Amazonia* [Richey, J., M. McClaine, and R. Victoria (eds.)]. pp. 17-42.
- 56 Martin, R.V., D.J. Jacob, J.A. Logan, I. Bey, R.M. Yantosca, A.C. Staudt, Q.B. Li, A.M. Fiore, B.N.  
57 Duncan, H. Liu, P. Ginoux, and V. Thouret, 2002: Interpretation of TOMS observations of tropical

- 1 tropospheric ozone with a global model and in-situ observations. *Journal of Geophysical Research*,  
2 107(D18), 4351, doi:10.1029/2001JD001480.
- 3 Martin, R.V., D.J. Jacob, R.M. Yantosca, M. Chin, and P. Ginoux, 2003a: Global and regional decreases in  
4 tropospheric oxidants from photochemical effects of aerosols. *Journal of Geophysical Research*,  
5 108(D3), 4097, doi:10.1029/2002JD002622. In newrefs.djj updated 1.doc this is referred to as 2003b.  
6 Need to verify!!!!
- 7 Martin, R.V., D.J. Jacob, K.V. Chance, T.P. Kurosu, P.I. Palmer, and M.J. Evans, 2003b: Global inventory  
8 of nitrogen oxide emissions constrained by space-based observations of NO<sub>2</sub> columns. *Journal of*  
9 *Geophysical Research*, 108 (D17), 4537, doi:10.1029/2003JD003453.
- 10 Martinerie, P., G.P. Brasseur, and C. Granier, 1995: The chemical composition of ancient atmospheres: a  
11 model study constrained by ice core data. *Journal of Geophysical Research*, 100, 14291-14304.
- 12 Matrai, P.A., and M.D. Keller, 1993: Dimethylsulphide in a large-scale coccolithophore bloom in the Gulf of  
13 Maine. *Continental Shelf Research*, 13(8-9), 831-843.
- 14 Matthews, R., and R. Wassmann, 2003: Modelling the impacts of climate change and methane emission  
15 reductions on rice production: a review. *European Journal of Agronomy*, 19, 573-598.
- 16 Matthews, H.D., A.J. Weaver, and K.J. Meissner, 2004a: Terrestrial carbon cycle dynamics under recent and  
17 future climate change. *Journal of Climate*, 18, 1609-1628.
- 18 Matthews, H.D., A.J. Weaver, K.J. Meissner, N.P. Gillett, and M. Eby, 2004b: Natural and anthropogenic  
19 climate change: incorporating historical land cover change, vegetation dynamics and the global carbon  
20 cycle. *Climate Dynamics*, 22(5), 461-479.
- 21 Matthews, H.D., M. Eby, A.J. Weaver, and B.J. Hawkins, 2005: Primary productivity control of simulated  
22 carbon cycle-climate feedbacks. *Geophysical Research Letters*, 32, L14708.
- 23 Maynard, K., and J.-F. Royer, 2004a: Sensitivity of a general circulation model to land surface parameters in  
24 African tropical deforestation experiments. *Climate Dynamics*, 22, doi:10.1007/s0038200403989.
- 25 Maynard, K., and J.-F. Royer, 2004b: Effects of "realistic" land-cover change on a greenhouse-warmed  
26 African climate. *Climate Dynamics*, 22, 343-358, doi:10.1007/s00382-003-0371-z.
- 27 McCabe, G.J., M.P. Clark, and M.C. Serreze, 2001: Trends in northern hemisphere surface cyclone  
28 frequency and intensity. *Journal of Climate*, 14, 2763-2768.
- 29 McFiggans, G.M., P. Artaxo, U. Baltensperger, H. Coe, M.C. Facchina, G. Feingold, S. Fuzzi, M. Gysel, A.  
30 Laaksonen, U. Lohmann, T.F. Mentel, D. Murphy, C.D. O'Dowd, J.R. Snider, and E. Weingartner,  
31 2005: The effect of aerosol composition and properties on warm cloud droplet activation. *Atmospheric*  
32 *Chemistry & Physics Discussions*, 5, (submitted).
- 33 McGuffie, K., and A. Henderson-Sellers, 2004: Stable water isotope characterization of human and natural  
34 impacts on land-atmosphere exchanges in the Amazon basin. *Journal of Geophysical Research*, 109,  
35 D17104, doi:10.1029/2003JD004388.
- 36 McKinley, G.A., M.J. Follows, and J. Marshall, 2004a: Mechanisms of air-sea CO<sub>2</sub> flux variability in the  
37 equatorial Pacific and the North Atlantic. *Global Biogeochemical Cycles*, 18, GB2011,  
38 doi:10.1029/2003GB002179.
- 39 McKinley, G.A., C. Rödenbeck, S. Houwelling, M. Gloor, and M. Heimann, 2004b: Pacific dominance to  
40 global air-sea CO<sub>2</sub> flux variability: A novel atmospheric inversion agrees with ocean models.  
41 *Geophysical Research Letters*, 31, L22308, doi:10.1029/2004GL021069.
- 42 McLinden, C., S. Olsen, B. Hannegan, O. Wild, M. Prather, and J. Sundet, 2000: Stratospheric ozone in 3-D  
43 models: a simple chemistry and the cross-tropopause flux. *Journal of Geophysical Research*, 105,  
44 14653-14665.
- 45 Medina, J., and A. Nenes, 2004: Effects of film-forming compounds on the growth of giant cloud  
46 condensation nuclei: implications of cloud microphysics and the aerosol indirect effect. *Journal of*  
47 *Geophysical Research*, 109, doi:10.1029/2004JD004666.
- 48 Meissner, K.J., A.J. Weaver, H.D. Matthews, and P.M. Cox, 2003: The role of land-surface dynamics in  
49 glacial inception: a study with the UVic Earth System Climate Model. *Climate Dynamics*, 21, 515-537.
- 50 Melillo, J.M., P.A. Steudler, J.D. Aber, K. Newkirk, H. Lux, F.P. Bowles, C. Catricala, A. Magill, T.  
51 Ahrens, and S. Morrisseau, 2002: Soil warming and carbon-cycle feedbacks to the climate system.  
52 *Science*, 298, 2173-2176.
- 53 Menon, S., and A.D.D. Genio, 2005: Human induced climate change: an interdisciplinary assessment,  
54 (submitted).
- 55 Menon, S., A.D. DelGenio, D. Koch, and G. Tselioudis, 2002a: GCM Simulations of the aerosol indirect  
56 effect: sensitivity to cloud parameterization and aerosol burden. *Journal of Atmospheric Science*, 59,  
57 692-713.

- 1 Menon, S., J. Hansen, L. Nazarenko, and Y. Luo, 2002b: Climate effects of black carbon aerosols in China  
2 and India. *Science*, 297, 2250-2252.
- 3 Meshkhidze, N., W.L. Chameides, A. Nenes, and G. Chen, 2003: Iron mobilization in mineral dust: can  
4 anthropogenic SO<sub>2</sub> emissions affect ocean productivity? *Geophysical Research Letters*, 30(21), 2085,  
5 doi:10.1029/2003GL018035.
- 6 Mickley, L.J., P.P. Murti, D.J. Jacob, J.A. Logan, D.M. Koch, and D. Rind, 1999: Radiative forcing from  
7 tropospheric ozone calculated with a unified chemistry-climate model. *Journal of Geophysical  
8 Research-Atmospheres*, 104(D23), 30153-30172.
- 9 Mickley, L.J., D.J. Jacob, and D. Rind, 2001: Uncertainty in preindustrial abundance of tropospheric ozone:  
10 implications for radiative forcing calculations. *Journal of Geophysical Research*, 106, 3389-3399.
- 11 Mickley, L.J., D.J. Jacob, B.D. Field, and D. Rind, 2004: Effects of future climate change on regional air  
12 pollution episodes in the United States. *Geophysical Research Letters*, 30, 1862,  
13 doi:10.1029/2003GL017933.
- 14 Mikaloff Fletcher, S.E., P.P. Tans, L.M. Bruhwiler, J.B. Miller, and M. Heimann, 2004a: CH<sub>4</sub> sources  
15 estimated from atmospheric observations of CH<sub>4</sub> and its <sup>13</sup>C/<sup>12</sup>C isotopic ratios: 1. Inverse modeling of  
16 source processes. *Global Biogeochemical Cycles*, 18, GB4004, doi:10.1029/2004GB002223.
- 17 Mikaloff Fletcher, S.E., P.P. Tans, L.M. Bruhwiler, J.B. Miller, and M. Heimann, 2004b: CH<sub>4</sub> sources  
18 estimated from atmospheric observations of CH<sub>4</sub> and its <sup>13</sup>C/<sup>12</sup>C isotopic ratios: 2. Inverse modeling of  
19 CH<sub>4</sub> fluxes from geographical regions. *Global Biogeochemical Cycles*, 18,  
20 doi:10.1029/2004GB002224.
- 21 Milkov, A.V., 2004: Global estimates of hydrate-bound gas in marine sediments: how much is really out  
22 there? *Earth-Science Reviews*, 66, 183-197.
- 23 Millero, F.J., D. Pierrot, K. Lee, R. Wanninkhof, R. Feely, C.L. Sabine, R.M. Key, and T. Takahashi, 2002:  
24 Dissociation constants for carbonic acid determined from field measurements. *Deep-Sea Research I*, 49,  
25 1705-1723.
- 26 Milly, P.C.D. and A.B. Schmakin, 2002a: Global modeling of land water and energy balances, Part I: The  
27 Land Dynamics (LaD) model. *Journal of Hydrometeorology*, 3, 301-310.
- 28 Milly, P.C.D. and A.B. Schmakin, 2002b: Global modeling of land water and energy balances, Part II: land-  
29 characteristic contributions to spatial variability. *Journal of Hydrometeorology*, 3, 301-310.
- 30 Ming, Y., V. Ramaswamy, P.A. Ginoux, L.H. Horowitz, and L.M. Russell, 2005: GFDL GCM simulations  
31 of the indirect radiative effects of anthropogenic aerosols, (submitted).
- 32 Mohanty, U., D.S. Niyogi, S. Raman, and A. Sarkar, 2001: Numerical study of the role of land-air-sea  
33 interactions for the northeasterly monsoon circulations over Indian Ocean during INDOEX. *Current  
34 Science*, 80 (Suppl. S), 60-68.
- 35 Monnin, E., A. Indermühle, A. Dallenbach, J. Fluckiger, B. Stauffer, T.F. Stocker, D. Raynaud, and J.M.  
36 Barnola, 2001: Atmospheric CO<sub>2</sub> concentrations over the last glacial termination. *Science*, 291(5501),  
37 112-114.
- 38 Moorcroft, P.R., G.C. Hurtt, and S.W. Pacala, 2001: A method for scaling vegetation dynamics: The  
39 ecosystem demography model (ED). *Ecological Monographs*, 71(4), 557-585, 2001.
- 40 Mopper, K., and E.T. Degens, 1979: Organic carbon in the ocean: nature and cycling. In: *The global carbon  
41 cycle* [Bolin, B., E.T. Degens, S. Kempe, and P. Ketner (eds.)]. John Wiley and Sons, 293-316.
- 42 Mudie, P.J., A. Rochon, and E. Levac, 2002: Palynological records of red-tide producing species in Canada:  
43 past trends and implications for the future. *Paleogeography Paleoclimatology Paleoecology*, 180(1-3),  
44 159-186.
- 45 Nadelhoffer, K., B.P. Colman, W.S. Currie, A.H. Magill, and J.D. Aber, 2004: Decadal-scale fates of N-15  
46 tracers added to oak and pine stands under ambient and elevated N inputs at the Harvard Forest (USA).  
47 *Forest Ecology and Management*, 196, 89-107.
- 48 Naja, M., H. Akimoto, and J. Staehelin, 2003: Ozone in background and photochemically aged air over  
49 central Europe: analysis of long-term ozonesonde data from Hohenpeissenberg and Payerne. *Journal of  
50 Geophysical Research*, 108(D2), 4063, doi:10.1029/2002JD002477.
- 51 Nakajima, T., A. Higurachi, K. Kawamoto, and J.E. Penner, 2001: A possible correlation between satellite-  
52 derived cloud and aerosol microphysical parameters. *Geophysical Research Letters*, 28, 1171-1174.
- 53 National Research Council [NRC], 1991: *Rethinking the Ozone Problem in Urban and Regional Air  
54 Pollution*. National Academy Press, Washington, DC.
- 55 Nemani, R., M. White, P. Thornton, K. Nishida, S. Reddy, J. Jenkins, and S. Running, 2002: Recent trends  
56 in hydrologic balance have enhanced the terrestrial carbon sink in the United States. *Geophysical  
57 Research Letters*, 29(10), 1468, doi:10.1029/2002GL014867.

- 1 Nemani, R.R., C.D. Keeling, H. Hashimoto, W.M. Jolly, S.C. Piper, C.J. Tucker, R.B. Myneni, and S.W.  
2 Running, 2003: Climate-driven increases in global terrestrial net primary production from 1982 to  
3 1999. *Science*, 300: 1560-1563.
- 4 Nenes, A., R.J. Charlson, M.C. Facchini, M. Kulmala, A. Laaksonen, and J.H. Seinfeld, 2002: Can chemical  
5 effects on cloud droplet number rival the first indirect effect? *Geophysical Research Letters*, 29,  
6 doi:10.1029/2002GL015295.
- 7 Nepstad, D., 2002: Forest fires: Behavior and ecological effects. *Nature*, 415(6871), 476-476.
- 8 Nepstad, D.C., P. Moutinho, M.B. Dias-Filho, E. Davidson, G. Cardinot, D. Markewitz, R. Figueiredo, N.  
9 Vianna, J. Chambers, D. Ray, J.B. Guerreiros, P. Lefebvre, L. Sternberg, M. Moreira, L. Barros, F.Y.  
10 Ishida, I. Tohlver, E. Belk, K. Kalif, and K. Schwalbe, 2002: The effect of partial throughfall exclusion  
11 on canopy processes and biogeochemistry of an Amazon forest. *Journal of Geophysical Research*,  
12 107(D20), 1-18.
- 13 Nepstad, D., P. Lefebvre, U.L. Da Silva, J. Tomasella, P. Schlesinger, L. Solorzano, P. Moutinho, D. Ray,  
14 and J.G. Benito, 2004: Amazon drought and its implications for forest flammability and tree growth: a  
15 basin-wide analysis. *Global Change Biology*, 10(5), 704-717.
- 16 Nesbitt, S.W., R.Y. Zhang, and R.E. Orville, 2000: Seasonal and global NO<sub>x</sub> production by lightning  
17 estimated from the Optical Transient Detector (OTD). *Tellus B*, 52, 1206-1215.
- 18 Newman, P.A., S.R. Kawa, and E.R. Nash, 2004: On the size of the Antarctic ozone hole. *Geophysical  
19 Research Letters*, 31, L21104, doi:10.1029/2004GL020596.
- 20 Niu, G.-Y., and Z.-L. Yang, 2004: Effects of vegetation canopy processes on snow surface energy and mass  
21 balances. *Journal of Geophysical Research*, 109, D23111, doi: 10.1029/2004JD004884.
- 22 Nober, F.J., H.-F. Graf, and D. Rosenfeld, 2003: Sensitivity of the global circulation to the suppression of  
23 precipitation by anthropogenic aerosols. *Global Planetary Change*, 37, 57-80.
- 24 Novelli, P.C., P.M. Lang, K.A. Masarie, D.F. Hurst, R. Myers, J.W., and J.W. Elkins, 1999: Molecular  
25 hydrogen in the troposphere: global distribution and budget. *Journal of Geophysical Research*,  
26 104(D23), 30427-30444.
- 27 O'Dowd, C.D., J.A. Lowe, and M.H. Smith, 1999: Coupling sea-salt and sulphate interactions and its impact  
28 on cloud droplet concentration predictions. *Geophysical Research Letters*, 26, 1311-1314.
- 29 O'Dowd, C.D., M.C. Facchina, F. Cavalli, D. Ceburnis, M. Mircea, S. Decesari, S. Fuzzi, Y.J. Yoon, and J.-  
30 P. Putaud, 2004: Biogenically driven organic contribution to marine aerosols. *Nature*, 431, 676-680.
- 31 Ogawa, K., and T. Schmugge, 2004: Mapping surface broadband emissivity of the sahara desert using  
32 ASTER and MODIS data. *Earth Interactions*, 8(7), 1-14.
- 33 Oglesby, R.J., S. Marshall, D.J. Erickson III, J.O. Roads, and F.R. Robertson, 2002: Thresholds in  
34 atmosphere-soil moisture interactions: results from climate model studies. *Journal of Geophysical  
35 Research*, 107(14), doi:10.1029/2001JD001045.
- 36 Oku, Y., and H. Ishikawa, 2004: Estimates of land temperature over the Tibetan plateau using GMS data.  
37 *Journal of Applied Meteorology*, 43, 548-561.
- 38 Oleson, K.W., G.B. Bonan, C. Schaaf, F. Gao, Y. Jin, and A. Strahler, 2003: Assessment of global climate  
39 model land surface albedo using MODIS data. *Geophysical Research Letters*, 30(8), 1443,  
40 doi:10.1029/2002GL016749.
- 41 Oleson, K.W., G.B. Bonan, S. Levis, and M. Vertenstein, 2004: Effects of land use change on North  
42 American climate: impact of surface datasets and model biogeophysics. *Climate Dynamics*, 23, 117-132  
43 doi:10.1007/s00382-004-0426-9.
- 44 Oleson, K.W., Y. Dai, G. Bonan, M. Bosilovich, R. Dickinson, P. Dirmeyer, F. Hoffman, P. Houser, S.  
45 Levis, G.-Y. Niu, P. Thornton, M. Vertenstein, Z.-L. Yang, X. Zeng, 2004: *Technical Description of the  
46 Community Land Model (CLM)*, NCAR Technical Note NCAR/TN-461+STR, National Center for  
47 Atmospheric Research, Boulder, Colorado, USA, 173 pp.
- 48 Ollinger, S.V., and J.D. Aber, 2002: The interactive effects of land use, carbon dioxide, ozone, and N  
49 deposition. *Global Change Biology*, 8, 545-562
- 50 Olsen, S.C., C.A. McLinden, and M.J. Prather, 2001: Stratospheric N<sub>2</sub>O-NO<sub>y</sub> system: testing uncertainties in  
51 a three-dimensional framework. *Journal of Geophysical Research*, 106, 28771-28784.
- 52 Oltmans, S. H., Vömel, D. Hofmann, K. Rosenlof, and D. Kley, 2000: The increase in stratospheric water  
53 vapor from balloonborne, frostpoint hygrometer measurements at Washington, D.C., and Boulder,  
54 Colorado. *Geophysical Research Letters*, 27 (21), 3453-3456.
- 55 O'Neill, K.P., E.S. Kasischke, and D.D. Richter, 2003: Seasonal and decadal patterns of soil carbon uptake  
56 and emission along an age sequence of burned black spruce stands in interior Alaska. *Journal of  
57 Geophysical Research-Atmospheres*, 108 (D1), 8155, doi:10.1029/2001JD000443.

- 1 Orcutt, K.M., F. Lipschultz, K. Gundersen, R. Arimoto, A.F. Michaels, A.H. Knap, and J.R. Gallon, 2001: A  
2 seasonal study of the significance of N<sub>2</sub> fixation by *Trichodesmium* spp. at the Bermuda Atlantic Time-  
3 series Study (BATS) site. *Deep-Sea Research II*, 48, 1583-1608.
- 4 Ordóñez, C., H. Mathis, M. Furger, S. Henne, C. Hüglin, J. Staehelin, and A.S.H. Prévôt, 2005: Changes of  
5 daily surface ozone maxima in Switzerland in all seasons from 1992 to 2002 and discussion of summer  
6 2003. *Atmospheric Chemistry & Physics*, 5, 1187-1203.
- 7 Oren, R., D.S. Ellsworth, K.H. Johnsen, N. Phillips, B.E. Ewers, C. Maier, K.V.R. Schafer, H. McCarthy, G.  
8 Hendrey, S.G. McNulty, and G.G. Katul, 2001: Soil fertility limits carbon sequestration by forest  
9 ecosystems in a CO<sub>2</sub>-enriched atmosphere. *Nature*, 411(6836), 469-472.
- 10 Orphan, V.J., C.H. House, K.-U. Hinrichs, K.D. McKeegan, and E.F. DeLong, 2002: Multiple archaeal  
11 groups mediate methane oxidation in anoxic cold seep sediments. *Proceedings of the National Academy  
12 of Sciences USA*, 99: 7663-7668.
- 13 Orr, J.C., 2002: *Global ocean storage of anthropogenic carbon (GOSAC), final report*. Environment Canada  
14 Environment and Climate Programme (Contranc ENV4-CT97-0495), IPSL/CNRS, France, 129 pp.  
15 [http://www.ipsl.jussieu.fr/OCMIP/reports/GOSAC\\_finalreport\\_hires.pdf](http://www.ipsl.jussieu.fr/OCMIP/reports/GOSAC_finalreport_hires.pdf),
- 16 Orr, J.C., E. Maier-Reimer, U. Mikolajewicz, P. Monfray, J.L. Sarmiento, J.R. Toggweiler, N.K. Taylor, J.  
17 Palmer, N. Gruber, C.L. Sabine, C. Le Quere, R.M. Key, and J. Boutin, 2001: Estimates of  
18 anthropogenic carbon uptake from four three-dimensional global ocean models. *Global Biogeochemical  
19 Cycles*, 15(1), 43-60, doi:10.1029/1999GB001256.
- 20 Orr, J.C., V.J. Fabry, O. Aumont, Laurent Bopp, S.C. Doney, R.M. Feely, A. Gnanadesikan, N. Gruber, A.  
21 Ishida, F. Joos, R.M. Key, K. Lindsay, E. Maier-Reimer, R. Matear, P. Monfray, A., Mouchet, R.G.  
22 Najjar, G.-K. Plattner, K.B. Rodgers, C.L. Sabine, J.L. Sarmiento, R. Schlitzer, R.D. Slater, I.J.  
23 Totterdell, M.-F. Weirig, Y. Yamanaka, and A. Yool, 2005: Decline in ocean carbonate and high-  
24 latitude aragonitic organisms. *Nature*, (in revision).
- 25 Osborne, T.M., D.M. Lawrence, J.M. Slingo, A.J. Challinor, and T.R. Wheeler, 2004: Influence of  
26 vegetation on the local climate and hydrology in the tropics: sensitivity to soil parameters. *Climate  
27 Dynamics*, 23, 45-61.
- 28 Osterkamp, T.E., and V.E. Romanovsky, 1999: Evidence for warming and thawing of discontinuous  
29 permafrost in Alaska. *Permafrost and Periglacial Processes*, 10(1), 17-37.
- 30 Oyama, M.D., and C.A. Nobre, 2004: Climatic consequences of a large-scale desertification in northeast  
31 Brazil: a GCM simulation study. *Journal of Climate*, 17(16), 3203-3213.
- 32 Pacala, S.W., G.C. Hurtt, D. Baker, P. Peylin, R.A. Houghton, R.A. Birdsey, L. Heath, E.T. Sundquist, R.F.  
33 Stallard, P. Ciais, P. Moorcroft, J.P. Caspersen, E. Shevliakova, B. Moore, G. Kohlmaier, E. Holland,  
34 M. Gloor, M.E. Harmon, S.M. Fan, J.L. Sarmiento, C.L. Goodale, D. Schimel, and C.B. Field, 2001:  
35 Consistent land- and atmosphere-based US carbon sink estimates. *Science*, 292, 2316-2320.
- 36 Page, S., F. Siegert, J.O. Rieley, H-D.V. Boehm, A. Jaya, and S. Limin, 2002: The amount of carbon released  
37 from peat and forest fires in Indonesia during 1997. *Nature*, 320, 61-65.
- 38 Palmer, P.I., D.J. Jacob, D.B.A. Jones, C.L. Heald, R.M. Yantosca, J.A. Logan, G.W. Sachse, and D.G.  
39 Streets, 2003a: Inverting for emissions of carbon monoxide from Asia using aircraft observations over  
40 the western Pacific. *Journal of Geophysical Research*, 108(D21), 8828, doi:10.1029/2003JD003397.
- 41 Palmer, P.I., D.J. Jacob, A.M. Fiore, R.V. Martin, K. Chance, and T. Kurosui, 2003b: Mapping isoprene  
42 emissions over North America using formaldehyde column observations from space. *Journal of  
43 Geophysical Research*, 108, 4180, doi:10.1029/2002JD002153.
- 44 Park, J., D. Jacob, P.I. Palmer, A.D. Clarke, R.J. Weber, M.A. Zondlo, F.L. Eisele, A. Bandy, D. Thornton,  
45 G. Sachse, and T.C. Bond, 2005: Export efficiency of black carbon aerosol in continental outflow:  
46 global implications. *Journal of Geophysical Research*, 110, D11205, doi:10.1029/2004JD005432.
- 47 Park, R.J., K.E. Pickering, D.J. Allen, G.L. Stenchikov, and M.S. Fox-Rabinovitz, 2004: Global simulation  
48 of tropospheric ozone using the University of Maryland Chemical Transport Model (UMD-CTM): 1.  
49 Model description and evaluation. *Journal of Geophysical Research*, 109, D09301,  
50 doi:10.1029/2003JD004266.
- 51 Passow, U., 2002: Transparent exopolymer particles (TEP) in aquatic environments. *Progress in  
52 Oceanography*, 55(3-4), 287-333.
- 53 Passow, U., R.F. Shipe, A. Murray, D.K. Pak, M.A. Brzezinski, and A.L. Alldredge, 2001: The origin of  
54 transparent exopolymer particles (TEP) and their role in the sedimentation of particulate matter.  
55 *Continental Shelf Research*, 21(4), 327-346.

- 1 Peng, Y., and U. Lohmann, 2003: Sensitivity study of the spectral dispersion of the cloud droplet size  
2 distribution on the indirect aerosol effect. *Geophysical Research Letters*, 30(10), 1507,  
3 doi:10.1029/2003GL017192.
- 4 Peng, Y., U. Lohmann, R. Leaitch, C. Banic, and M. Couture, 2002: The cloud albedo-cloud droplet  
5 effective radius relationship for clean and polluted clouds from ACE and FIRE. *Journal of Geophysical*  
6 *Research*, 107(D11), doi:10.1029/2002JD000281.
- 7 Penner, J.E., H. Eddleman, and T. Novakov, 1993: Towards the development of a global inventory of black  
8 carbon emissions. *Atmospheric Environment*, 27A, 1277-1295.
- 9 Penner, J., M. Andreae, H. Annegram, L. Barrie, J. Feichter, D. Hegg, A. Jayaraman, R. Leaitch, D. Murphy,  
10 J. Nganga, and G. Pitari, 2001: Aerosols, their direct and indirect effects. In: *Climate Change 2001: The*  
11 *Scientific basis*. Cambridge University Press, 289-348.
- 12 Penner, J.E., S.Y. Zhang, and C.C. Chuang, 2003: Soot and smoke aerosol may not warm climate. *Journal of*  
13 *Geophysical Research*, 108(21), 4657, doi:10.1029/2003JD003409.
- 14 Penner, J.E., X. Dong, and Y. Chen, 2004: Observational evidence of a change in radiative forcing due to the  
15 indirect aerosol effect. *Nature*, 427, 231-234.
- 16 Pereira, J.M.C., 2003: Remote sensing of burned areas in tropical savannas. *International Journal of*  
17 *Wildland Fire*, 12(3-4), 259-270.
- 18 Perlwitz, J., I. Tegen, and R.L. Miller, 2001: Interactive soil dust aerosol model in the GISS GCM 1.  
19 Sensitivity of the soil dust cycle to radiative properties of soil dust aerosols. *Journal of Geophysical*  
20 *Research*, 106(D16), doi:10.1029/2000JD900668.
- 21 Peters, W., M. Krol, F. Dentener, and J. Lelieveld, 2001: Identification of an El Niño-southern oscillation  
22 signal in a multiyear global simulation of tropospheric ozone. *Journal of Geophysical Research*,  
23 106(D10), doi:10.1029/2000JD900658.
- 24 Petit, J., J. Jouzel, D. Raynaud, N. Barkov, J.-M. Barnola, I. Basile, M. Bender, J. Chappellaz, M. Davis, G.  
25 Delaygue, M. Delmotte, V. Kotlyakov, M. Legrand, V. Lipenkov, C. Lorius, L. Pepin, C. Ritz, E.  
26 Saltzman, and M. Stievenard, 1999: Climate and atmospheric history of the past 420,000 years from the  
27 Vostok ice core, Antarctica. *Nature*, 399, 429-436.
- 28 Pétron, G., C. Granier, B. Khattatov, V. Yudin, J.-F. Lamarque, L. Emmons, J. Gille, and D.P. Edwards,  
29 2004: Monthly CO surface sources inventory based on the 2000-2001 MOPITT data. *Geophysical*  
30 *Research Letters*, 31, L21107. doi:10.1029/2004GL020560.
- 31 Peylin, P., P. Bousquet, C. Le Quéré, S. Stith, P. Friedlingstein, G. McKinley, N. Gruber, P. Rayner, and P.  
32 Ciais, 2005: Multiple constraints on regional CO<sub>2</sub> flux variations over land and oceans. *Global*  
33 *Biogeochemical Cycles*, 19, GB1011, doi: 10.1029/2003GB002214.
- 34 Phillips, O.L., Y. Malhi, N. Higuchi, W.F. Laurance, P.V. Núñez, R.M. Vásquez, S.G. Laurance, L.V.  
35 Ferreira, M. Stern, S. Brown, and J. Grace, 1998: Changes in the Carbon Balance of Tropical Forests:  
36 evidence from Long-Term Plots. *Science*, 282(5388), 439-442.
- 37 Phillips, V.T.J., T.W. Choullarton, A.M. Blyth, and J. Latham, 2002: The influence of aerosol concentrations  
38 on the glaciation and precipitation of a cumulus cloud. *Quarterly Journal of the Royal Meteorological*  
39 *Society*, 128(581), 951-971.
- 40 Pickering, K.E., A.M. Thompson, H. Kim, A.J. DeCaria, L. Pfister, T.L. Kucsera, J.C. Witte, M.A. Avery,  
41 D.R. Blake, J.H. Crawford, B.G. Heikes, G.W. Sachse, S.T. Sandholm, and R.W. Talbot, 2001: Trace  
42 gas transport and scavenging in PEM-Tropics B South Pacific convergence zone convection. *Journal of*  
43 *Geophysical Research*, 106(D23), doi:10.1029/2001JD000328.
- 44 Pielke, R.A., 2001: Influence of the spatial distribution of vegetation and soils on the prediction of cumulus  
45 convective rainfall. *Reviews of Geophysics*, 39(2), 151-177.
- 46 Pinker, R.T., B. Zhang, and E.G. Dutton, 2005: Do satellites detect trends in surface solar radiation? *Science*,  
47 308, 850-854.
- 48 Pinto, J. P., and M.A.K. Khalil, 1991: The stability of tropospheric OH during ice ages, inter-glacial epochs  
49 and moder times. *Tellus*, 43B, 347-352.
- 50 Pitman, A.J., B.J. McAvaney, N. Bagnound, and B. Chemint, 2004: Are inter-model differences in AMIP-II  
51 near surface air temperature means and extremes explained by land surface energy balance complexity.  
52 *Geophysical Research Letters*, 31, L05205, doi:10.1029/2003GL019233.
- 53 Platnick, S., P.A. Durkee, K. Nielsen, J.P. Taylor, S.-C. Tsay, M.D. King, R.J. Ferek, P.V. Hobbs, and J.W.  
54 Rottman, 2000: The role of background cloud microphysics in the radiative formation of ship tracks.  
55 *Journal of Atmospheric Science*, 57, 2607-2624.
- 56 Plattner, G.-K., F. Joos, T.F. Stocker, and O. Marchal, 2001: Feedback mechanisms and sensitivities of  
57 ocean carbon uptake under global warming. *Tellus Series B*, 53, 564-592.

- 1 Prather, M.J., 1996: Time scales in atmospheric chemistry: theory, GWPs for CH<sub>4</sub> and CO, and runaway  
2 growth. *Geophysical Research Letters*, 23(19), doi:10.1029/96GL02371.
- 3 Prather, M.J., 1998: Time scales in atmospheric chemistry: coupled perturbations to N<sub>2</sub>O, NO<sub>y</sub>, and O<sub>3</sub>.  
4 *Science*, 270, 1339-1341.
- 5 Prather, M.J., 2002: Lifetimes of atmospheric species: integrating environmental impacts. *Geophysical*  
6 *Research Letters*, 29(22), 2063, doi:10.1029/2002GL016299.
- 7 Prather, M., D. Ehhalt, F. Dentener, R.G. Derwent, E. Dlugokencky, E. Holland, I.S.A. Isaksen, J. Katima,  
8 V. Kirchhoff, P. Matson, P.M. Midgley, and M. Wang, 2001: Atmospheric chemistry and greenhouse  
9 gases. In: *Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third*  
10 *Assessment Report of the Intergovernmental Panel on Climate Change (IPCC)*, [Houghton, J. T., Y.  
11 Ding, D.J. Griggs, M. Noguer, P.J. van der Linden, X. Dai, K. Maskell, and C.A. Johnson (eds.)].  
12 Cambridge University Press, Cambridge, pp. 239–287.
- 13 Price, C., and D. Rind, 1994a: Possible implications of global climate change on global lightning  
14 distributions and frequencies. *Journal of Geophysical Research*, 99(D5), doi:10.1029/94JD00019.
- 15 Price, C., and D. Rind, 1994b: The impact of a 2-x-CO<sub>2</sub> climate on lightning-caused fires. *Journal of*  
16 *Climate*, 7, 1484-1494.
- 17 Price, C., J. Penner, and M. Prather, 1997: NO<sub>x</sub> from lightning 1. Global distribution based on lightning  
18 physics. *Journal of Geophysical Research*, 102(D5), doi:10.1029/96JD03504.
- 19 Prinn, R.G., J. Huang, R.F. Weiss, D.M. Cunnold, P.J. Fraser, P.G. Simmonds, A. McCulloch, C. Harth, P.  
20 Salameh, S. O'Doherty, R.H.J. Wang, L. Porter, and B.R. Miller, 2001: Evidence for substantial  
21 variations of atmospheric hydroxyl radicals in the past two decades. *Science*, 292(5523), 1882-1888.
- 22 Prinn, R.G., J. Huang, R.F. Weiss, D.M. Cunnold, P.J. Fraser, P.G. Simmonds, A. McCulloch, C. Harth, S.  
23 Reimann, P. Salameh, S.O'Doherty, R.H.J. Wang, L.W. Porter, B.R. Miller, and P.B. Krummel, 2005:  
24 Evidence for variability of atmospheric hydroxyl radicals over the past quarter century. *Geophysical*  
25 *Research Letters*, (submitted).
- 26 Prospero, J.M., P. Ginoux, O. Torres, S.E. Nicholson, and T.E. Gill, 2002: Environmental characterization of  
27 global sources of atmospheric soil dust identified with the NIMBUS 7 total ozone mapping  
28 spectrometer (TOMS) absorbing aerosol product. *Reviews of Geophysics*, 40,  
29 doi:10.1029/2000RG000095.
- 30 Qian, Y., and F. Giorgi, 2000: Regional climatic effects of anthropogenic aerosols? The case of  
31 Southwestern China. *Geophysical Research Letters*, 27(21), doi:10.1029/2000GL011942.
- 32 Quaas, J., O. Boucher and F.-M. Bréon, 2004: Aerosol indirect effects in POLDER satellite data and the  
33 Laboratoire de Météorologie Dynamique-Zoom (LMDZ) general circulation model. *Journal of*  
34 *Geophysical Research*, 109, doi:10.1029/2003JD004317.
- 35 Quideau, S.A., O.A. Chadwick, S.E. Trumbore, J.L. Johnson-Maynard, R.C. Graham, and M.A. Anderson,  
36 2001: Vegetation control on soil organic matter dynamics. *Organic Geochemistry*, 32(2), 247-252.
- 37 Rabalais, N.N., 2002: Nitrogen in aquatic ecosystems. *Ambio*, 31(2), 102-112.
- 38 Raddatz, T., J. Jungclaus, J. Kattge, W. Knorr, C. Reick, E. Roeckner, K.G. Schnitzler, R. Schnur, and P.  
39 Wetzell, 2005: Is the tropical land biosphere dominating the carbon cycle - climate feedback within the  
40 21st century? *Geophysical Research Letters*, (submitted).
- 41 Raich J., and W. Schlesinger, 1992: The global carbon dioxide flux in soil respiration and its relationship to  
42 vegetation and climate. *Tellus B*, 44, 81-99.
- 43 Ramanathan, V., C. Chung, D. Kim, T. Bettge, L. Buja, J.T. Kiehl, W.M. Washington, Q. Fu, D.R. Sikka,  
44 and M. Wild, 2005: Atmospheric brown clouds: impacts on South Asian climate and hydrological  
45 cycle. *Proceeding of the National Academy of Sciences USA*, 102, 5326-5333.
- 46 Ramanathan, V., P.J. Crutzen, J. Lelieveld, A.P. Mitra, D. Althausen, J. Anderson, M.O. Andreae, W.  
47 Cantrell, G.R. Cass, C.E. Chung, A.D. Clarke, J.A. Coakley, W.D. Collins, W.C. Conant, F. Dulac, J.  
48 Heintzenberg, A.J. Heymsfield, B. Holben, S. Howell, J. Hudson, A. Jayaraman, J.T. Kiehl, T.N.  
49 Krishnamurti, D. Lubin, G. McFarquhar, T. Novakov, J.A. Ogren, I.A. Podgorny, K. Prather, K.  
50 Priestley, J.M. Prospero, P.K. Quinn, K. Rajeev, P. Rasch, S. Rupert, R. Sadourny, S.K. Satheesh, G.E.  
51 Shaw, P. Sheridan, and F.P.J. Valero, 2001b: Indian Ocean Experiment: an integrated analysis of the  
52 climate forcing and effects of the great Indo-Asian haze. *Journal of Geophysical Research*, 106(D22),  
53 doi:10.1029/2001JD900133.
- 54 Ramanathan, V., P.J. Crutzen, J.T. Kiehl, and D. Rosenfeld, 2001a: Aerosols, climate, and the hydrological  
55 cycle. *Science*, 294, 2119-2123.

- 1 Randel, W.J., F. Wu, S.J. Oltmans, K. Rosenlof, and G.E. Nedoluha, 2004: Interannual changes of  
2 stratospheric water vapor and correlations with tropical tropopause temperatures. *Journal of*  
3 *Atmospheric Science*, 61, 2133-2148.
- 4 Randerson, J.T., F.S. Chapin, J.W. Harden, J.C. Neff, and M.E. Harmon, 2002a: Net ecosystem production:  
5 A comprehensive measure of net carbon accumulation by ecosystems. *Ecological Applications*, 12(4),  
6 937-947.
- 7 Randerson, J.T., I.G. Enting, E.A.G. Schuur, K. Caldeira, and I.Y. Fung, 2002b: Seasonal and latitudinal  
8 variability of troposphere Delta(CO<sub>2</sub>)-C-14: Post bomb contributions from fossil fuels, oceans, the  
9 stratosphere, and the terrestrial biosphere. *Global Biogeochemical Cycles*, 16(4), 1112,  
10 doi:10.1029/2002GB001876.
- 11 Randerson, J.T., G.J. Collatz, J.E. Fessenden, A.D. Munoz, C.J. Still, J.A. Berry, I.Y. Fung, N. Suits, and  
12 A.S. Denning, 2002c: A possible global covariance between terrestrial gross primary production and C-  
13 13 discrimination: Consequences for the atmospheric C-13 budget and its response to ENSO. *Global*  
14 *Biogeochemical Cycles*, 16(4), 1136, doi:10.1029/2001GB001845.
- 15 Randerson, J.T., C.J. Still, J.J. Balle, I.Y. Fung, S.C. Doney, P.P. Tans, T.J. Conway, J.W.C. White, B.  
16 Vaughn, N. Suits, and A.S. Denning, 2002d: Carbon isotope discrimination of arctic and boreal biomes  
17 inferred from remote atmospheric measurements and a biosphere-atmosphere model. *Global*  
18 *Biogeochemical Cycles*, 16(3), doi:10.1029/2001GB001435.
- 19 Randerson, J.T., G.R. van der Werf, G.J. Collatz, L. Giglio, C.J. Still, P. Kasibhatla, J.B. Miller, J.W.C.  
20 White, R.S. DeFries, and E.S. Kasischke, 2005: Fire emissions from C-3 and C-4 vegetation and their  
21 influence on interannual variability of atmospheric CO<sub>2</sub> and delta(CO<sub>2</sub>)-C-13. *Global Biogeochemical*  
22 *Cycles*, 19(2), GB2019, doi:10.1029/2004GB002366.
- 23 Raven, J.A., and P.G. Falkowski, 1999: Oceanic sinks for atmospheric CO<sub>2</sub>. *Plant Cell and Environment*,  
24 22(6), 741-755.
- 25 Raymond, P.A., and J.J. Cole, 2003: Increase in the export of alkalinity from North America's largest river.  
26 *Science*, 301, 88-91.
- 27 Rayner, P.J., I.G. Enting, R.J. Francey, and R. Langenfelds, 1999: Reconstructing the recent carbon cycle  
28 from atmospheric CO<sub>2</sub>, δ<sup>13</sup>C and O<sub>2</sub>/N<sub>2</sub> observations. *Tellus Series B-Chemical and Physical*  
29 *Meteorology*, 51(2), 213-232.
- 30 Reale, O., and P. Dirmeyer, 2002: Modeling the effect of land surface evaporation variability on  
31 precipitation variability. I: General response. *Journal of Hydrometeorology*, 3(4), 433-450.
- 32 Reale, O., P. Dirmeyer, and A. Schlosser, 2002: Modeling the effect of land surface evaporation variability  
33 on precipitation variability. II: Time- and space-scale structure. *Journal of Hydrometeorology*, 3(4),  
34 451-466.
- 35 Reichenau, T.G., and G. Esser, 2003: Is interannual fluctuation of atmospheric CO<sub>2</sub> dominated by combined  
36 effects of ENSO and volcanic aerosols? *Global Biogeochemical Cycles*, 17(4), 1094,  
37 doi:10.1029/2002GB002025.
- 38 Rice, A.H., E.H. Pyle, S.R. Saleska, L.H., M. Palace, M. Keller, P.B. de Camargo, K. Portilho, D.F.  
39 Marques, and S.C. Wofsy, 2004: Carbon balance and vegetation dynamics in an old-growth Amazonian  
40 Forest. *Ecological Applications*, 14(4), S55-S71 (Suppl. S).
- 41 Richter, A., and J.P. Burrows, 2002: Tropospheric NO<sub>2</sub> from GOME measurements. *Advances in Space*  
42 *Research*, 29, 1673-1683.
- 43 Richter, A., J.P. Burrows, H. Nuss, C. Granier, and U. Niemeier, 2005: Increase in tropospheric nitrogen  
44 dioxide over China observed from space. *Nature*, 437, 129-132.
- 45 Richter, D.D., D. Markewitz, S.E. Trumbore, and C.G. Wells, 1999: Rapid accumulation and turnover of soil  
46 carbon in a re-establishing forest. *Nature*, 400(6739), 56-58.
- 47 Ridgwell, A.J., S.J. Marshall, and K. Gregson, 1999: Consumption of atmospheric methane by soils: A  
48 process-based model. *Global Biogeochemical Cycles*, 13, 59-70.
- 49 Riebesell, U., D.A. Wolf-Gladrow, and V. Smetacek, 1993: Carbon dioxide limitation of marine  
50 phytoplankton growth rates. *Nature*, 361, 249-251.
- 51 Riebesell, U., I. Zondervan, B. Rost, P.D. Tortell, R.E. Zeebe, and F.M.M. Morel, 2000: Reduced  
52 calcification of marine plankton in response to increased atmospheric CO<sub>2</sub>. *Nature*, 407, 364-367.
- 53 Rind, D., J. Lerner, and C. McLinden, 2001: Changes of tracer distribution in the doubled CO<sub>2</sub> climate.  
54 *Journal of Geophysical Research*, 106(D22), doi:10.1029/2001JD000439.
- 55 Rinke, A., K. Dethloff, and M. Fortmann, 2004: Regional climate effects of Arctic Haze. *Geophysical*  
56 *Research Letters*, 31, L16202, doi:10.1029/2004GL020318.

- 1 Roberts, D.L., and A. Jones, 2004: Climate sensitivity to black carbon aerosol from fossil fuel combustion.  
2 *Journal of Geophysical Research*, 109, D16202, doi:10.1029/2004JD004676.
- 3 Robock, A., 2001: Stratospheric forcing needed for dynamical seasonal prediction. *Bulletin of the American*  
4 *Meteorological Society*, 82, 2189-2192.
- 5 Rodell, M., P.R. Houser, U. Jambor, J. Gottschalk, K. Mitchell, C.-J. Meng, K. Arsenault, B. Cosgrove, J.  
6 Radakovich, M. Bosilovich, J.K. Entin, J.P. Walker, D. Lohmann, and D. Toll, 2004: The global land  
7 data assimilation system. *Bulletin of the American Meteorological Society*, 85(3), 381-394.
- 8 Rödenbeck, C., S. Houweling, M. Gloor, and M. Heimann, 2003: CO<sub>2</sub> flux history 1982–2001 inferred from  
9 atmospheric data using a global inversion of atmospheric transport. *Atmospheric Chemistry & Physics*,  
10 3, 2575-2659.
- 11 Roderick, M.L., and G.D. Farquhar, 2002: The cause of decreased pan evaporation over the past 50 years.  
12 *Science*, 298, 1410-1411.
- 13 Roderick, M.L., G.D. Farquhar, S.L. Berry, and I.R. Noble, 2001: On the direct effect of clouds and  
14 atmospheric particles on the productivity and structure of vegetation. *Oecologia*, 129, 21-30.
- 15 Roeckner, E., L. Bengtsson, J. Feichter, J. Lelieveld, and H. Rodhe, 1999: Transient climate change  
16 simulations with a coupled atmosphere-ocean GCM including the tropospheric sulphur cycle. *Journal*  
17 *of Climate*, 12, 3004-3032.
- 18 Roeckner, E., P. Stier, J. Feichter, S. Kloster, and M. Esch, 2005: Impact of carbonaceous aerosol forcing on  
19 regional climate change. *Journal of Climate*, (submitted).
- 20 Rokich, D.P., K.W. Dixon, K. Sivasithamparam, and K.A. Meney, 2002. Smoke, mulch, and seed  
21 broadcasting effects on woodland restoration in Western Australia. *Restoration Ecology*, 10(2), 185-  
22 194.
- 23 Rolim, S.G., R.M. Jesus, H.E.M. Nascimento, H.T.Z. do Couto, and J.Q. Chambers, 2005: Biomass change  
24 in an Atlantic tropical moist forest: the ENSO effect in permanent sample plots over a 22-year period.  
25 *Oecologia*, 142(2), 238-246.
- 26 Rosenfeld, D. and W.L. Woodley, 2000: Deep convective clouds with sustained supercooled liquid water  
27 down to -37.5 °C. *Nature*, 405, 440-442.
- 28 Rosenfeld, D., 1999: TRMM observed first direct evidence of smoke from forest fires inhibiting rainfall.  
29 *Geophysical Research Letters*, 26(20), doi:10.1029/1999GL006066.
- 30 Rosenfeld, D., 2000: Suppression of rain and snow by urban and industrial air pollution. *Science*, 287, 1793-  
31 1796.
- 32 Rosenfeld, D., and G. Feingold, 2003: Explanation of discrepancies among satellite observations of the  
33 aerosol indirect effects. *Geophysical Research Letters*, 30(14), 1776, doi:10.1029/2003GL017684.
- 34 Rosenfeld, D., Y. Rudich, and R. Lahav, 2001: Desert dust suppressing precipitation: a possible  
35 desertification feedback loop. *Proceedings of the National Academy of Sciences USA*, 98, 5975-5980.
- 36 Rosenfeld, D., R. Lahav, A. Khain, and M. Pinsky, 2002: The role of sea spray in cleansing air pollution  
37 over ocean via cloud processes. *Science*, 297, 1667-1670.
- 38 Rosenfeld, J.E., A.R. Douglass, and D.B. Considine, 2002: The impact of increasing carbon dioxide on  
39 ozone recovery. *Journal of Geophysical Research*, 107(D6), 4049, doi:10.1029/2001JD000824.
- 40 Rosenlof, K.H., S.J. Oltmans, D. Kley, J.M. Russell, E.W. Chiou, W.P. Chu, D.G. Johnson, K.K. Kelly,  
41 H.A. Michelsen, G.E. Nedoluha, E.E. Remsberg, G.C. Toon, and M.P. McCormick, 2001: Stratospheric  
42 water vapor increases over the past half-century. *Geophysical Research Letters*, 28, 1195-1198.
- 43 Rotman, D.A., C.S. Atherton, D.J. Bergmann, P.J. Cameron-Smith, C.C. Chuang, P.S. Connell, J.E. Dignon,  
44 A. Franz, K.E. Grant, D.E. Kinnison, C.R. Molenkamp, D.D. Proctor, and J.R. Tannahill, 2004:  
45 IMPACT, the LLNL 3-D global atmospheric chemical transport model for the combined troposphere  
46 and stratosphere: Model description and analysis of ozone and other trace gases. *Journal of Geophysical*  
47 *Research*, 109(D4), D04303, doi:10.1029/2002JD003155.
- 48 Rotstayn, L.D., and Y. Liu, 2003: Sensitivity of the first indirect aerosol effect to an increase of cloud  
49 droplet spectral dispersion with droplet number concentration. *Journal of Climate*, 16, 3476-3481.
- 50 Rotstayn, L.D., and Y. Liu, 2005: A smaller global estimate of the second indirect aerosol effect.  
51 *Geophysical Research Letters*, 32, L05708, doi:10.1029/2004GL021922.
- 52 Rotstayn, L., and U. Lohmann, 2002: Tropical rainfall trends and the indirect aerosol effect. *Journal of*  
53 *Climate*, 15, 2103-2116.
- 54 Roy, S.B., G.C. Hurtt, C.P. Weaver, and S.W. Pacala, 2003: Impact of historical land cover change on the  
55 July climate of the United States. *Journal of Geophysical Research*, 108(D24), 4793,  
56 doi:10.1029/2003JD003565.

- 1 Roy, T., P. Rayner, R. Matear, and R. Francey, 2003: Southern hemisphere ocean CO<sub>2</sub> uptake: reconciling  
2 atmospheric and oceanic estimates. *Tellus Series B*, 55(2), 701-710.
- 3 Royal Society, 2005: *Ocean acidification due to increasing atmospheric carbon dioxide*. The Royal Society,  
4 London, United Kingdom, Policy document 12/05, June 2005, ISBN 0854036172, 60pp.  
5 <http://www.royalsoc.ac.uk/document.asp?tip=0&id=3249>
- 6 Russell, J.L., and J.M. Wallace, 2004: Annual carbon dioxide drawdown and the Northern Annular Mode,  
7 *Global Biogeochemical Cycles*, 18(1), GB1012, doi:10.1029/2003GB002044.
- 8 Sabine, C.L., R.M. Key, K.M. Johnson, F.J. Millero, A. Poisson, J.L. Sarmiento, D.W.R. Wallace, and C.D.  
9 Winn, 1999: Anthropogenic CO<sub>2</sub> inventory of the Indian Ocean. *Global Biogeochemical Cycles*, 13(1),  
10 doi:10.1029/1998GB900022.
- 11 Sabine, C.L., R.A. Feely, R.M. Key, J.L. Bullister, F.J. Millero, K. Lee, T.H. Peng, B. Tilbrook, T. Ono, and  
12 C.S. Wong, 2002: Distribution of anthropogenic CO<sub>2</sub> in the Pacific Ocean. *Global Biogeochemical*  
13 *Cycles*, 16(4), 1083, doi:10.1029/2001GB001639.
- 14 Sabine, C.L., R.A. Feely, N. Gruber, R.M. Key, K. Lee, J.L. Bullister, R. Wanninkhof, C.S. Wong, D.W.R.  
15 Wallace, B. Tilbrook, F.J. Millero, T.H. Peng, A. Kozyr, T. Ono, and A.F. Rios, 2004: The oceanic sink  
16 for anthropogenic CO<sub>2</sub>. *Science*, 305(5682), 367-371.
- 17 Sanderson, M.G., W.J. Collins, R.G. Derwent, and C.E. Johnson, 2003a: Simulation of global hydrogen  
18 levels using a lagrangian three-dimensional model. *Journal of Atmospheric Chemistry*, 46(1), 15-28.
- 19 Sanderson, M.G., C.D. Jones, W.J. Collins, C.E. Johnson, and R.G. Derwent, 2003b: Effect of climate  
20 change on isoprene emissions and surface ozone levels. *Geophysical Research Letters*, 30(18), 1936,  
21 doi:10.1029/2003GL017642.
- 22 Sarmiento, J.L., and E.T. Sundquist, 1992: Revised budget for the oceanic uptake of anthropogenic carbon  
23 dioxide, *Nature*, 356, 589-593.
- 24 Sarmiento, J.L., R. Slater, R. Barber, L. Bopp, S.C. Doney, A.C. Hirst, J. Kleypas, R. Matear, U.  
25 Mikolajewicz, P. Monfray, V. Soldatov, S.A. Spall, and R. Stouffer, 2004: Response of ocean  
26 ecosystems to climate warming. *Global Biogeochemical Cycles*, 18(3), GB3003,  
27 doi:10.1029/2003GB002134.
- 28 Sass, R.L., J.A. Andrews, A.J. Ding, and F.M. Fisher, 2002: Spatial and temporal variability in methane  
29 emissions from rice paddies: implications for assessing regional methane budgets. *Nutrient Cycling in*  
30 *Agroecosystems*, 64 (1-2), 3-7.
- 31 Satheesh, S.K., 2002: Aerosol radiative forcing over land: effect of surface and cloud reflection. *Annales*  
32 *Geophysicae*, 20(12), 2105-2109.
- 33 Satheesh, S.K., and V. Ramanathan, 2000: Large differences in tropical aerosol forcing at the top of the  
34 atmosphere and Earth's surface. *Nature*, 405, doi:10.1038/35011039.
- 35 Scanlon, B.R., D.G. Levitt, R.C. Reedy, K.E. Keese, and M.J. Sully, 2005: Ecological controls on water-  
36 cycle response to climate variability in deserts. *Proceedings of the National Academy of Sciences USA*,  
37 102(17), 6033-6038. [www.pnas.org/cgi/doi/10.1073/pnas.0408571102](http://www.pnas.org/cgi/doi/10.1073/pnas.0408571102)
- 38 Schaap, M., H.A.C.D. Van Der Gon, F.J. Dentener, A.J.H. Visschedijk, M. Van Loon, H.M. ten Brink, J.-P.  
39 Putaud, B. Guillaume, C. Liousse, and P.J.H. Builtjes, 2004: Anthropogenic black carbon and fine  
40 aerosol distribution over Europe. *Journal of Geophysical Research*, 109, D18207,  
41 doi:10.1029/2003JD004330.
- 42 Schimel, D.S., J.I. House, K.A. Hibbard, P. Bousquet, P. Ciais, P. Peylin, B.H. Braswell, M.J. Apps, D.  
43 Baker, A. Bondeau, J. Canadell, G. Churkina, W. Cramer, A.S. Denning, C.B. Field, P. Friedlingstein,  
44 C. Goodale, M. Heimann, R.A. Houghton, J.M. Melillo, B. Moore III, D. Murdiyarso, I. Noble, S.W.,  
45 Pacala, I.C. Prentice, M.R. Raupach, P.J. Rayner, R.J. Scholes, W.L. Steffen, and C. Wirth, 2001:  
46 Recent patterns and mechanisms of carbon exchange by terrestrial ecosystems. *Nature*, 414, 169-172.
- 47 Schoeberl, M.R., A.R. Douglass, Z. Zhu, and S. Pawson, 2003: A comparison of the lower stratospheric age-  
48 spectra derived from a general circulation model and two data assimilation systems. *Journal of*  
49 *Geophysical Research*, 108(D3), doi:10.1029/2002JD002652.
- 50 Schultz, M.G., T. Diehl, G.P. Brasseur, and W. Zittel, 2003: Air pollution and climate-forcing impacts of a  
51 global hydrogen economy. *Science*, 302, 624-627.
- 52 Schwartz, M.D., and T.R. Karl, 1990: Spring phenology – Nature's experiment to detect the effect of green-  
53 up on surface maximum temperatures. *Monthly Weather Review*, 118(4), 883-890.
- 54 Schwartz R.D., 2005: Global dimming: clear-sky atmospheric transmission from astronomical extinction  
55 measurements. *Journal of Geophysical Research*, 110, D14210, doi:10.1029/2005JD005882.
- 56 Schwartz, S.E., 1993: Does fossil fuel combustion lead to global warming? *Energy*, 18(12), 1229-1248.

- 1 Sciandra, A., J. Harlay, D. Lefèvre, R. Lemée, P. Rimmelin, M. Denis, and J.-P. Gattuso, 2003: Response of  
2 the coccolithophorid *Emiliania huxleyi* to elevated partial pressure of CO<sub>2</sub> under nitrogen limitation.  
3 *Marine Ecology Progress Series*, 261, 111-122.
- 4 Seifert, A., and K.D. Beheng, 2005: A two-moment cloud microphysics parameterization for mixed-phase  
5 clouds. Part II: Maritime vs. continental deep convective storms. *Meteorology and Atmospheric*  
6 *Physics*, (in press).
- 7 Seiler, W., and R. Conrad, 1987: Contribution of tropical ecosystems to the global budget of trace gases,  
8 especially CH<sub>4</sub>, H<sub>2</sub>, CO, and N<sub>2</sub>O, In: *The Geophysiology of Amazonia: Vegetation and Climate*  
9 *Interactions*, [Dickerson, R.E. (ed.)]. John Wiley, New York, pp. 33-62.
- 10 Sharma, S., D. Lavoué, H. Cachier, L.A. Barrie, and S.L. Gong, 2004: Long-term trends of the black carbon  
11 concentrations in the Canadian Arctic. *Journal of Geophysical Research*, 109, D15203,  
12 doi:10.1029/2003JD004331.
- 13 Shaw, G.E., 1983: Bio-controlled thermostasis involving the sulphur cycle. *Climate Change*, 5, 297-303.
- 14 Sherwood, S., 2002: A microphysical connection among biomass burning, cumulus clouds, and stratospheric  
15 moisture. *Science*, 295, 1272-1275.
- 16 Shim, C., Y.H. Wang, Y. Choi, P.I. Palmer, D.S. Abbot, and K. Chance, 2005: Constraining global isoprene  
17 emissions with GOME formaldehyde column measurements. *Journal of Geophysical Research*, (in  
18 press).
- 19 Shindell, D.T., and G. Faluvegi, 2002: An exploration of ozone changes and their radiative forcing prior to  
20 the chlorofluorocarbon era. *Atmospheric Chemistry & Physics*, 2, 363-374.
- 21 Shindell, D.T., J.L. Grenfell, D. Rind, V. Grewe, and C. Price, 2001: Chemistry–climate interactions in the  
22 Goddard Institute for Space Studies general circulation model 1. Tropospheric chemistry model  
23 description and evaluation. *Journal of Geophysical Research-Atmospheres*, 106(D8),  
24 doi:10.1029/2000JD900704.
- 25 Shindell, D.T., G. Faluvegi, and N. Bell, 2003: Preindustrial-to-present-day radiative forcing by tropospheric  
26 ozone from improved simulations with the GISS chemistry-climate GCM. *Atmospheric Chemistry &*  
27 *Physics*, 3, 1675-1702.
- 28 Shindell, D.T., B.P. Walter, and G. Faluvegi, 2004: Impacts of climate change on methane emissions from  
29 wetlands. *Geophysical Research Letters*, 31, L21202, doi:10.1029/2004GL021009.
- 30 Shugart, H.H., N.H.F. French, E.S. Kasischke, J.J. Slawski, C.W. Dull, R.A. Shuchman, and J. Mwangi,  
31 2001: Detection of vegetation change using reconnaissance imagery. *Global Change Biology*, 7(3), 247-  
32 252.
- 33 Sievering, H., I. Fernandez, J. Lee, J. Horn, and L. Rustad, 2000: Forest canopy uptake of atmospheric  
34 nitrogen deposition at eastern U.S. conifer sites: Carbon storage implications. *Global Biogeochemical*  
35 *Cycles*, 14(4), doi:10.1029/1999GB001250.
- 36 Sigman, D.M., and E.A. Boyle, 2000: Glacial/interglacial variations in atmospheric carbon dioxide. *Nature*,  
37 407, 859-869.
- 38 Sillman, S., and P.J. Samson, 1995: Impact of temperature on oxidant photochemistry in urban, polluted  
39 rural, and remote environments. *Journal of Geophysical Research*, 100(D6), 11497-11508,  
40 doi:10.1029/94JD02146.
- 41 Silva Dias, M.A.F., S. Rutledge, P. Kabat, P.L. Silva Dias, C. Nobre, G. Fisch, A.J. Dolman, E. Zipser, M.  
42 Garstang, A. Manzi, J.D. Fuentes, H. Rocha, J. Marengo, A. Plana-Fattori, L. Sá, R. Alvalá, M.O.  
43 Andreae, P. Artaxo, R. Gielow, L. Gatti, 2002: Clouds and rain processes in a biosphere atmosphere  
44 interaction context. *Journal of Geophysical Research*, 107(D20), 8072, doi:10.1029/2001JD000335.
- 45 Simmonds, P.G., R.G. Derwent, S. O'Doherty, D.B. Ryall, L.P. Steele, R.L. Langenfelds, P. Salameh, H.J.  
46 Wang, C.H. Dimmer, and L.E. Hudson, 2000: Continuous high-frequency observations of hydrogen at  
47 the Mace Head baseline atmospheric monitoring station over the 1994-1998 period. *Journal of*  
48 *Geophysical Research*, 105(D10), 12105-12121, doi:10.1029/2000JD900007.
- 49 Simpson, I.J., D.R. Blake, and F.S. Rowland, 2002: Implications of the recent fluctuation in the growth rate  
50 of tropospheric methane. *Geophysical Research Letters*, 29(10), 1479, doi:10.1029/2001GL014521.
- 51 Singh, H.B., L.J. Salas, R.B. Chatfield, E. Czech, A. Fried, J. Walega, M.J. Evans, B.D. Field, D.J. Jacob, D.  
52 Blake, B. Heikes, R. Talbot, G. Sachse, J.H. Crawford, M.A. Avery, S. Sandholm, and H. Fuelberg,  
53 2004: Analysis of the atmospheric distribution, sources, and sinks of oxygenated volatile organic  
54 chemicals based on measurements over the Pacific during TRACE-P. *Journal of Geophysical Research*,  
55 109, D15S07, doi:10.1029/2003JD003883.
- 56 Sinha, A., 1995: Relative influence of lapse rate and water vapour on the greenhouse effect. *Journal of*  
57 *Geophysical Research*, 100(D3), 5095-5103, doi:10.1029/94JD03248.

- 1 Sitch, S., B. Smith, I.C. Prentice, A. Arneth, A. Bondeau, W. Cramer, J.O. Kaplan, S. Levis, W. Lucht, M.T.  
2 Sykes, K. Thonicke, and S. Venevsky, 2003: Evaluation of ecosystem dynamics, plant geography and  
3 terrestrial carbon cycling in the LPJ dynamic global vegetation model. *Global Change Biology*, 9, 161-  
4 185.
- 5 Sitch, S., V. Brovkin, W. von Bloh, D. van Vuuren, B. Eickhout, and A. Ganopolski, 2005: Impacts of future  
6 land cover changes on atmospheric CO<sub>2</sub> and climate. *Global Biogeochemical Cycles*, 19,  
7 doi:10.1029/2004GB002311.
- 8 Smith, K.A., and F. Conen, 2004: Impacts of land management on fluxes of trace greenhouse gases. *Soil Use  
9 And Management*, 20, 255-263.
- 10 Smith, N.V., S.S. Saatchi, and J.T. Randerson, 2004: Trends in high northern latitude soil freeze and thaw  
11 cycles from 1988 to 2002. *Journal of Geophysical Research-Atmospheres*, 109(D12), D12101,  
12 doi:10.1029/2003JD004472.
- 13 Smith, S.V., and J.T. Hollibaugh, 1993: Coastal metabolism and the oceanic organic carbon balance.  
14 *Reviews of Geophysics*, 31(1), 75-89.
- 15 Smyth, T.J., T. Tyrrell, and B. Tarrant, 2004: Time series coccolithophore activity in the Barents Sea, from  
16 twenty years of satellite imagery. *Geophysical Research Letters*, 31, L11302,  
17 doi:10.1029/2004GL019735.
- 18 Snyder, P.K., C. Delire, and J.A. Foley, 2004: Evaluating the influence of different vegetation biomes on the  
19 global climate. *Climate Dynamics*, 23, 279-302, doi: 10.1007/s00382-004-0430-0.
- 20 Snyder, P.K., J.A. Foley, M.H. Hitchman, and C. Delire, 2004: Analyzing the effects of complete tropical  
21 forest removal on the regional climate using a detailed three-dimensional energy budget: an application  
22 to Africa. *Journal of Geophysical Research*, 109, D21102, doi:10.1029/2003JD004462.
- 23 Sokolik, I.N., and O.B. Toon, 1996: Direct radiative forcing by anthropogenic airborne mineral aerosols.  
24 *Nature*, 381, 681-683.
- 25 Sombroek, W.G., P.M. Fearnside, and M. Cravo, 2000: Geographic assessment of carbon stored in  
26 Amazonian terrestrial ecosystems and their soils in particular. In: *Global climate change and tropical  
27 ecosystems* [Lal, R., J.M. Kimble, and B.A. Stewart (eds.)]. Boca Raton, FL, USA: CRC Press, pp. 375-  
28 389.
- 29 Stanhill, G., and S. Cohen, 2001: Global dimming a review of the evidence for a widespread and significant  
30 reduction in global radiation with discussion of its probable causes and possible agricultural  
31 consequences. *Journal of Agricultural and Forest Meteorology*, 107, 255-278.
- 32 Steele, L.P., E.J. Dlugokencky, P.M. Lang, P.P. Tans, R.C. Martin, and K.A. Masarie, 1992: Slowing down  
33 of the global accumulation of atmospheric methane during the 1980s. *Nature*, 358, 313-316.
- 34 Steinkamp, R., W. Zimmer, and H. Papen. 2001. Improved method for detection of methanotrophic bacteria  
35 in forest soils by PCR. *Current Microbiology*, 42, 316-322.
- 36 Stephens, G.L., N.B. Wood, and L.A. Pakula, 2004: On the radiative effects of dust on tropical convection.  
37 *Geophysical Research Letters*, 31, L23112, doi:10.1029/2004GL021342.
- 38 Stevenson, D.S., C.E. Johnson, W.J. Collins, R.G. Derwent, and J.M. Edwards, 2000: Future estimates of  
39 tropospheric ozone radiative forcing and methane turnover - the impact of climate change. *Geophysical  
40 Research Letters*, 105(14), doi:10.1029/1999GL010887.
- 41 Stevenson, D.S., R.M. Doherty, M.G. Sanderson, W.J. Collins, C.E. Johnson, and R.G. Derwent, 2004:  
42 Radiative forcing from aircraft NO<sub>x</sub> emissions: mechanisms and seasonal dependence. *Journal of  
43 Geophysical Research*, 109, D17307, doi:10.1029/2004JD004759.
- 44 Stevenson, D.S., F.J. Dentener, M.G. Schultz, K. Ellingsen, T.P.C. van Noije, O. Wild, G. Zeng, M. Amann,  
45 C.S. Atherton, N. Bell, D.J. Bergmann, I. Bey, T. Butler, J. Cofala, W.J. Collins, R.G. Derwent, R.M.  
46 Doherty, J. Drevet, H.J. Eskes, A.M. Fiore, M. Gauss, D.A. Hauglustaine, L.W. Horowitz, I.S.A.  
47 Isaksen, M.C. Krol, J.-F. Lamarque, M.G. Lawrence, V. Montanaro, J.-F. Müller, G. Pitari, M.J. Prather,  
48 J.A. Pyle, S. Rast, J.M. Rodriguez, M.G. Sanderson, N.H. Savage, D.T. Shindell, S.E. Strahan, K. Sudo,  
49 and S. Szopa, 2005a: Multi-model ensemble of present-day and near-future tropospheric ozone. *Journal  
50 of Geophysical Research*, (submitted).
- 51 Stevenson, D.S., R.M. Doherty, M.G. Sanderson, C.E. Johnson, W.J. Collins, and R.G. Derwent, 2005b:  
52 Impacts of climate change and variability on tropospheric ozone and its precursors. *Faraday  
53 Discussions*, 130, doi:10.1039/b417412g.
- 54 Stieglitz, M., A. Ducharne, R. Koster, and M. Suarez, 2001: The impact of detailed snow physics on the  
55 simulation of snow cover and subsurface thermodynamics at continental scales. *Journal of  
56 Hydrometeorology*, 2, 228-242.

- 1 Stieglitz, M., S.J. Dery, V.E. Romanovsky, and T.E. Osterkamp, 2003: The role of snow cover in the  
2 warming of arctic permafrost. *Geophysical Research Letters*, 30(13), 1721, doi:10.1029.2003GL017337.
- 3 Stier, P., J. Feichter, S. Kloster, E. Vignati, and J. Wilson, 2005: Emission-induced nonlinearities in the  
4 global aerosol system - results from the ECHAM5-HAM aerosol-climate model. *Journal of Climate*, 18,  
5 (submitted).
- 6 Stocks, B.J., M.A. Fosberg, T.J. Lynham, L. Mearns, B.M. Wotton, Q. Yang, J.Z. Jin, K. Lawrence, G.R.  
7 Hartley, J.A. Mason, and D.W. McKenney, 1998: Climate change and forest fire potential in Russian  
8 and Canadian boreal forests. *Climatic Change*, 38, 1-13.
- 9 Storelvmo, T., J.E. Kristjansson, S.J. Ghan, A. Kirkevag, and O. Seland, 2005: Predicting cloud droplet  
10 number concentration in CAM-Oslo. *Journal of Geophysical Research*, 110, (submitted).
- 11 Strack, J.E., G.E. Liston, and R.A. Pielke Sr., 2004: Modeling snow depth for improved simulation of snow-  
12 vegetation-atmosphere interactions. *Journal of Hydrometeorology*, 5(5), 723-734.
- 13 Stuber, N., M. Ponater, and R. Sausen, 2001: Is the climate sensitivity to ozone perturbations enhanced by  
14 stratospheric water vapor feedback? *Geophysical Research Letters*, 28(15),  
15 doi:10.1029/2001GL013000.
- 16 Sturm, M., J. Holmgren, J.P. McFadden, G.E. Liston, F.S. Chapin III, and C.H. Racine, 2001: Snow-shrub  
17 interactions in Arctic Tundra: a hypothesis with climatic implications. *Journal of Climate*, 14(3), 336-  
18 344.
- 19 Sudo, K., and M. Takahashi, 2001: Simulation of tropospheric ozone changes during 1997–1998 El Niño:  
20 meteorological impact on tropospheric photochemistry. *Geophysical Research Letters*, 28(21),  
21 doi:10.1029/2001GL013335.
- 22 Sukhinin, A.I., N.H.F. French, E.S. Kasischke, J.H. Hewson, A.J. SoJa, I.A. Csiszar, E.J. Hyer, T. Loboda,  
23 S.G. Conard, V.I. Romasko, E.A. Pavlichenko, S.I. Miskiva, and O.A. Slinkina, 2005: AVHRR-based  
24 mapping of fires in Russia: New products for fire management and carbon cycle studies (vol 93, pg 546,  
25 2004). *Remote Sensing of Environment*, 94(3), 428-428.
- 26 Sundquist, E.T., 1993: The global carbon dioxide budget, *Science*, 259(5097), 934-941.
- 27 Suzuki, K., T. Nakajima, A. Numaguti, T. Takemura, K. Kawamoto, and A. Higurashi, 2004: A study of the  
28 aerosol effect on a cloud field with simultaneous use of GCM modeling and satellite observations.  
29 *Journal of Atmospheric Science*, 61, 179-194.
- 30 Takahashi, T., S.C. Sutherland, C. Sweeney, A. Poisson, N. Metzl, B. Tilbrook, N. Bates, R. Wanninkhof,  
31 R.A. Feely, C. Sabine, J. Olafsson, and Y. Nojiri, 2002: Global sea-air CO<sub>2</sub> flux based on climatological  
32 surface ocean pCO<sub>2</sub>, and seasonal biological and temperature effects. *Deep-Sea Research II*, 49(9-10),  
33 1601-1622.
- 34 Takahashi, T., S.C. Sutherland, R.A. Feely, and C.E. Cosca, 2003: Decadal variation of the surface water  
35 PCO<sub>2</sub> in the Western and Central Equatorial Pacific. *Science*, 302, 852-856.
- 36 Tan, W.W., M.A. Geller, S. Pawson, and A. da Silva, 2004: A case study of excessive subtropical transport  
37 in the stratosphere of a data assimilation system. *Journal of Geophysical Research*, 109, D11102,  
38 doi:10.1029/2003JD004057.
- 39 Tans, P., and T. Conway, 1999: Development of the CO<sub>2</sub> latitude gradient in recent decades. *Global*  
40 *Biogeochemical Cycles*, 13, 821-826.
- 41 Taylor, J.A., and J. Lloyd, 1992: Sources and sinks of atmospheric CO<sub>2</sub>. *Australian Journal of Botany*, 40,  
42 407-418.
- 43 Tegen, I., A.A. Lacis, and I. Fung, 1996: The influence of mineral aerosols from disturbed soils on the global  
44 radiation budget. *Nature*, 380, 419-422.
- 45 Tegen, I., S.P. Harrison, K. Kohfeld, I.C. Prentice, M. Coe, and M. Heimann, 2002. Impact of vegetation and  
46 preferential source areas on global dust aerosol: results from a model study. *Journal of Geophysical*  
47 *Research*, 107(D21), 4576. doi:10.1029/2001JD000963.
- 48 Tegen, I., M. Werner, S.P. Harrison, and K.E. Kohfeld, 2004: Relative importance of climate and land use in  
49 determining present and future global soil dust emission. *Geophysical Research Letters*, 31, L05105,  
50 doi:10.1029/2003GL019216.
- 51 Telles, E.D.C., P.B. de Camargo, L.A. Martinelli, S.E. Trumbore, E.S. da Costa, J. Santos, N. Higuchi, and  
52 R.C. Oliveira, 2003: Influence of soil texture on carbon dynamics and storage potential in tropical forest  
53 soils of Amazonia. *Global Biogeochemical Cycles*, 17(2), 1040, doi:10.1029/2002GB001953.
- 54 Textor, C., M. Schulz, S. Guibert, S. Kinne, S.E. Bauer, Y. Balkanski, T. Berntsen, T. Berglen, O. Boucher,  
55 M. Chin, F. Dentener, T. Diehl, H. Feichter, D. Fillmore, S. Ghan, P. Ginoux, S. Gong, A. Grini, J.  
56 Hendricks, L. Horowitz, I. Isaksen, T. Iversen, A. Kirkevag, D. Koch, J.E. Kristjansson, M. Krol, A.  
57 Lauer, J.F. Lamarque, X. Liu, V. Montanaro, G. Myhre, J. Penner, G. Pitari, S. Reddy, O. Seland, P.

- 1 Stier, T. Takemura, and X.Tie, 2005: Analysis and quantification of the diversities of aerosol life cycles  
2 within AEROCOM. *Atmospheric Chemistry & Physics Discussions*, (submitted).
- 3 Thomas, H., Y. Bozec, K. Elkalay, and H. de Baar, 2004: Enhanced open ocean storage of CO<sub>2</sub> from shelf  
4 sea pumping. *Science*, 304, 1005-1008.
- 5 Thompson, A.M., B.G. Doddridge, J.C. Witte, R.D. Hudson, W.T. Luke, J.E. Johnson, B.J. Johnson, S.J.  
6 Oltmans, and R. Weller, 2000: A tropical Atlantic paradox: shipboard and satellite views of a  
7 tropospheric ozone maximum and wave-one in January–February 1999. *Geophysical Research Letters*,  
8 27(20), 10.1029/1999GL011273.
- 9 Thompson, A.M., K.E. Pickering, D.P. Menamara, M.R. Schoeberl, R.D. Hudson, J.H. Kim, E.V. Browell,  
10 V.W.J.H. Kirchoff, and D. Nganga, 1996: Where did tropospheric ozone over southern Africa and the  
11 tropical Atlantic come from in October 1992? Insights from TOMS, GTE TRACE A, and SAFARI  
12 1992. *Journal of Geophysical Research*, 101(D19), doi:10.1029/96JD01463.
- 13 Thompson, S.L., B. Govindasamy, A. Mirin, K. Caldeira, C. Delire, J. Milovich, and M. Wickett, 2004:  
14 Quantifying the effects of CO<sub>2</sub>-fertilized vegetation on future global climate and carbon dynamics.  
15 *Geophysical Research Letters*, 31, L23211, doi:10.1029/2004GL021239.
- 16 Tian, H., J.M. Melillo, D.W. Kicklighter, A.D. McGuire, J.V.K. Helfrich III, B. Moore III, and C.J.  
17 Vororsmarty, 1998: Effect of interannual climate variability on carbon storage in Amazonian  
18 ecosystems. *Nature*, 396, 664–667.
- 19 Tian, Y., R.E. Dickinson, L. Zhou, R.B. Myneni, M. Friedl, C.B. Schaaf, M. Carroll, and F. Gao, 2004a:  
20 Land boundary conditions from MODIS data and consequences for the albedo of a climate model.  
21 *Geophysical Research Letters*, 31, L05504, doi:10.1029/2003GL019104.
- 22 Tian, Y., R.E. Dickinson, L. Zhou, X. Zeng, Y. Dai, R.B. Myneni, Y. Knyazikhin, X., Zhang, M. Friedl, H.  
23 Yu, W. Wu, and M. Shaikh, 2004b: Comparison of seasonal and spatial variations of leaf area index and  
24 fraction of absorbed photosynthetically active radiation from Moderate Resolution Imaging  
25 Spectroradiometer (MODIS) and common land model. *Journal of Geophysical Research*, 109,  
26 doi:10.1029/2003JD003777.
- 27 Tian, Y., R.E. Dickinson, L. Zhou, and M. Shaikh, 2004c: Impact of new land boundary conditions from  
28 Moderate Resolution Imaging Spectroradiometer (MODIS) data on the climatology of land surface  
29 variables. *Journal of Geophysical Research*, 109, D20115, doi:10.1029/2003JD004499.
- 30 Tie, X.X., A. Guenther, and E. Holland, 2003: Biogenic methanol and its impact on tropospheric oxidants.  
31 *Geophysical Research Letters*, 30(17), 1881, doi:10.1029/2003GL017167.
- 32 Tie, X.X., R.Y. Zhang, G. Brasseur, and W.F. Lei, 2002: Global NO<sub>x</sub> production by lightning. *Journal of*  
33 *Atmospheric Chemistry*, 43(1), 61-74.
- 34 Tie, X.X., S. Madronich, S. Walters, D.P. Edwards, P. Ginouz, N. Mahowald, R.Y. Zhang, C. Lou, and G.P.  
35 Brasseur, 2005: Assessment of the global impact of aerosols on tropospheric oxidants. *Journal of*  
36 *Geophysical Research*, 110, D03204, doi:10.1029/2004JD005359.
- 37 Timmreck, C., and M. Schulz, 2004: Significant dust simulation differences in nudged and climatological  
38 operation mode of the AGCM ECHAM. *Journal of Geophysical Research*, 109, D13202,  
39 doi:10.1029/2003JD004381.
- 40 Toggweiler, J.R., K. Dixon, and K. Bryan, 1989: Simulation of radiocarbon in a coarse resolution world  
41 ocean model, I. steady state pre-bomb distributions. *Journal of Geophysical Research*, 94, 8217-8242.
- 42 Torn, M.S., S.E. Trumbore, O.A. Chadwick, P.M. Vitousek, and D.M. Hendricks, 1997: Mineral control of  
43 soil organic carbon storage and turnover. *Nature*, 389(6647), 170-173.
- 44 Tortell, P.D., G.R. DiTullio, D.M. Sigman, and F.M.M. Morel, 2002: CO<sub>2</sub> effects on taxonomic composition  
45 and nutrient utilization in an Equatorial Pacific phytoplankton assemblage. *Marine Ecology Progress*  
46 *Series*, 236, 37-43.
- 47 Toumi, R., J.D. Haigh, and K.S. Law, 1996: A tropospheric ozone-lightning climate feedback. *Geophysical*  
48 *Research Letters*, 23(9), doi:10.1029/96GL00944.
- 49 Townsend, A.R., R.W. Howarth, M.S. Booth, C.C. Cleveland, S.K. Collinge, A.P. Dobson, P.R. Epstein,  
50 E.A. Holland, D.R. Keeny, and M.A. Malin, 2003: Human health effects of a changing global nitrogen  
51 cycle. *Frontiers in Ecology*, 1(5), 240-246.
- 52 Treseder, K.K., L.M. Egerton-Warburton, M.F. Allen, Y.F. Cheng, and W.C. Oechel, 2003: Alteration of  
53 soil carbon pools and communities of mycorrhizal fungi in chaparral exposed to elevated carbon  
54 dioxide. *Ecosystems*, 6(8), 786-796.
- 55 Tromp, T.K., R.-L. Shia, M. Allen, J.M. Eiler, and Y.L. Yung, 2003: Potential environmental impact of a  
56 hydrogen economy on the stratosphere. *Science*, 300, 1740-1742.

- 1 Trumbore, S.E., 1997: Potential responses of soil organic carbon to global environmental change.  
2 *Proceedings of the National Academy of Sciences of the USA*, 94(16), 8284-8291.
- 3 Trumbore, S.E., 2000: Age of soil organic matter and soil respiration: Radiocarbon constraints on  
4 belowground C dynamics. *Ecological Applications*, 10(2), 399-411.
- 5 Trumbore, S.E., and J.W. Harden, 1997: Accumulation and turnover of carbon in organic and mineral soils  
6 of the BOREAS northern study area. *Journal of Geophysical Research-Atmospheres*, 102(D24),  
7 doi:10.1029/97JD02231.
- 8 Trumbore, S.E., and J.B. Gaudinski, 2003: The secret lives of roots. *Science*, 302(5649), 1344-1345.
- 9 Trumbore, S.E., J.L. Bubier, J.W. Harden, and P.M. Crill, 1999: Carbon cycling in boreal wetlands: A  
10 comparison of three approaches. *Journal of Geophysical Research-Atmospheres*, 104(D22),  
11 doi:10.1029/1999JD900433.
- 12 Tsigaridis, K., and M. Kanakidou, 2003: Global modelling of secondary organic aerosol in the troposphere: a  
13 sensitivity analysis. *Atmospheric Chemistry & Physics*, 3, 1849-1869.
- 14 Tsvetsinskaya, E.A., C.B. Schaaf, F. Gao, A.H. Strahler, R.E. Dickinson, X. Zeng, and W. Lucht, 2002:  
15 Relating MODIS-derived surface albedo to soils and rock types over Northern Africa and the Arabia  
16 Peninsula. *Geophysical Research Letters*, 29(9), 10.1029/2001GLO14096.
- 17 Turner, D.P., J.V. Baglio, A.G. Wones, D. Pross, R. Vong, B.D. McVeety, and D.L. Phillips, 1991: Climate  
18 change and Isoprene emissions from vegetation. *Chemosphere*, 23, 37-56.
- 19 Turner, S.M., M.J. Harvey, C.S. Law, P.D. Nightingale, and P.S. Liss, 2004: Iron-induced changes in  
20 oceanic sulphur biogeochemistry. *Geophysical Research Letters*, 31(14), L14307,  
21 doi:10.1029/2004GL020296.
- 22 Tyrrell, T., P.M. Holligan, and C.D. Mobley, 1999: Optical impacts of oceanic coccolithophore blooms.  
23 *Journal of Geophysical Research*, 104(C2), 3223-3241.
- 24 Valentine, D.L., D.C. Blanton, W.S. Reeburgh, and M. Kastner, 2001: Water column methane oxidation  
25 adjacent to an area of active hydrate dissociation, Eel River Basin. *Geochimica et Cosmochimica Acta*,  
26 65, 2633-2640.
- 27 Van den Heever, S.C., G.G. Carrio, W.R. Cotton, P.J. DeMott, and A.J. Prenni, 2005: Impacts of nucleating  
28 aerosol on Florida convection, Part I: Mesoscale simulations. *Journal of Atmospheric Science*  
29 (submitted).
- 30 van den Hurk, B.J.J.M., P. Viterbo, and S.O. Los, 2003: Impact of leaf area index seasonality on the annual  
31 land surface evaporation in a global circulation model. *Geophysical Research Letters*, 108(D6), 4191,  
32 doi:10.1029/2002JD002846.
- 33 van der Werf, G., J.T. Randerson, G.J. Collatz, and L. Giglio, 2003: Carbon emissions from fires in tropical  
34 and subtropical ecosystems. *Global Change Biology*, 9(4), 547-562.
- 35 van der Werf, G.R., J.T. Randerson, G.J. Collatz, L. Giglio, P.S. Kasibhatla, A.F. Arellano Jr., S.C. Olsen,  
36 and E.S. Kasischke, 2004: Continental-scale partitioning of fire emissions during the 1997 to 2001 El  
37 niño/La niña period. *Science*, 303(5654), 73-76.
- 38 Van Dolah, F.M., 2000: Marine algae toxins: origins, health effects, and their increased occurrence.  
39 *Environmental Health Perspectives*, 108, 133-141 (Suppl. 1, Mar 2000).
- 40 van Noije, T.C.P., H.J. Eskes, M. Van Weele, and P.F.J. van Velthoven, 2004: Implications of enhanced  
41 Brewer-Dobson circulation in European Centre for Medium-Range Weather Forecasts reanalysis for the  
42 stratosphere-troposphere exchange of ozone in global chemistry transport models. *Journal of*  
43 *Geophysical Research*, 109, D19308, doi:10.1029/2004JD004586.
- 44 Velders, G.J.M., S. Madronich, C. Clerbaux, R. Derwent, M. Grutter, D. Hauglustaine, S. Incekik, M. Ko, J.-  
45 M. Libre, O. Nielsen, F. Stordal, and T. Zhu, 2005: Chemical and radiative effects of halocarbons and  
46 their replacement compounds. In: *Special Report on Safeguarding the Ozone Layer and Global Climate*  
47 *System*. IPCC/TEAP, Cambridge UK, pp. Chapter 2.
- 48 Veldkamp, E., A. Becker, L. Schwendenmann, D.A. Clark, and H. Schulte-Bisping, 2003: Substantial labile  
49 carbon stocks and microbial activity in deeply weathered soils below a tropical wet forest. *Global*  
50 *Change Biology*, 9, 1171-1184.
- 51 Vitousek, P., 2004: *Nutrient cycling and limitations: Hawai'i as a model ecosystem*. Princeton University  
52 Press, 232 pp.
- 53 Vitousek, P.M., J.D. Aber, R.W. Howarth, G.E. Likens, P.A. Matson, D.W. Schindler, W.H. Schlesinger,  
54 and D. Tilman, 1997: Human alteration of the global nitrogen cycle: sources and consequences.  
55 *Ecological Applications*, 7, 737-750.

- 1 Vitousek, P.M., L.O. Edin, P.A. Matson, J.H. Fownes, and J. Neff, 1998: Within-system element cycles,  
2 input-output budgets, and nutrient limitations. In: *Successes, Limitations, and Frontiers in Ecosystem*  
3 *Science* [Pace, M. and P. Groffman (eds.)]. Springer-Verlag, New York. pp. 432-451.
- 4 Voltaire, A., and J.-F. Royer, 2004: Tropical deforestation and climate variability. *Climate Dynamics*, 22,  
5 857-874. doi:10.1007/s00382-004-0423-z.
- 6 Von Kuhlmann, R., M.G. Lawrence, P.J. Crutzen, and P.J. Rasch, 2003: A model for studies of tropospheric  
7 ozone and nonmethane hydrocarbons: model description and ozone results. *Journal of Geophysical*  
8 *Research*, 108, 4294, doi:10.1029/2002JD002893.
- 9 Walsh, J.J., 1991: Importance of continental margins in the marine biogeochemical cycling of carbon and  
10 nitrogen. *Nature*, 350, 53-55.
- 11 Walter, B.P., and M. Heimann, 2000: A process-based, climate-sensitive model to derive methane emission  
12 from natural wetlands: application to five wetland sites, sensitivity to model parameters, and climate.  
13 *Global Biogeochemical Cycles*, 14, 745-765.
- 14 Walter, B.P., and M. Heimann, 2001a: Modeling modern methane emission from natural wetlands, 1. Model  
15 description and results. *Journal of Geophysical Research*, 106, 34189-34206.
- 16 Walter, B.P., and M. Heimann, 2001b: Modeling modern methane emission from natural wetlands, 2.  
17 Interannual variations 1982-1993. *Journal of Geophysical Research*, 106, 34207-37219.
- 18 Wang, C., 2004: A modeling study on the climate impacts of black carbon aerosols. *Journal of Geophysical*  
19 *Research*, 109, doi:10.1029/2003JD004084.
- 20 Wang, C., and R. Prinn, 1999: Impact of emissions, chemistry and climate on atmospheric carbon monoxide:  
21 100 year predictions from a global chemistry model. *Chemosphere (Global Change)*, 1, 73-81.
- 22 Wang, G., and E. Eltahir, 2000: Modeling the biosphere-atmosphere system: the impact of the subgrid  
23 variability in rainfall interception. *Journal of Climate*, 13, 2887-3078.
- 24 Wang, G., E.A.B. Eltahir, J.A. Foley, D. Pollard, and S. Levis, 2004: Decadal variability of rainfall in the  
25 Sahel: results from the coupled GENESIS-IBIS atmosphere-biosphere model. *Climate Dynamics*, 22,  
26 doi:10.1007/s00382-004-0411-3.
- 27 Wang, H., G.Y. Shi, T. Aoki, B. Wang, and T.L. Zhao, 2004: Radiative forcing due to dust aerosol over east  
28 Asia-north Pacific region during spring, 2001. *Chinese Science Bulletin*, 49, 1993-2000.
- 29 Wang, J., S., J.A. Logan, M.B. McElroy, B.N. Duncan, I.A. Megretskaya, and R.M. Yantosca, 2004: A 3-D  
30 model analysis of the slowdown and interannual variability in the methane growth rate from 1988 to  
31 1997. *Global Biogeochemical Cycles*, 18, GB3011, doi:10.1029/3003GB002180.
- 32 Wang, Y., D.J. Jacob, and J.A. Logan, 1998: Global simulation of tropospheric O<sub>3</sub>-NO<sub>x</sub>-hydrocarbon  
33 chemistry, 3. Origin of tropospheric ozone and effects of non-methane hydrocarbons. *Journal of*  
34 *Geophysical Research*, 103(D9), 10757-10768.
- 35 Wang, Y., R. Amundson, and S. Trumbore, 1999: The impact of land use change on C turnover in soils.  
36 *Global Biogeochemical Cycles*, 13(1), 47-57.
- 37 Wang, Z., X. Zeng, M. Barlage, R.E. Dickinson, F. Gao, and C.B. Schaaf, 2004: Using MODIS BRDF and albedo  
38 data to evaluate global model land surface albedo. *Journal of Hydrometeorology*, 5, 3-14.
- 39 Wanninkhof, R., and W.R. McGillis, 1999: A cubic relationship between air-sea CO<sub>2</sub> exchange and wind  
40 speed. *Geophysical Research Letters*, 26(13), 1889-1892.
- 41 Waring, R.H., and B.E. Law, 2001: The ponderosa pine ecosystem and environmental stress: Past, present  
42 and future. *Tree Physiology*, 21(5), 273-274.
- 43 Waring, R.H., and N. McDowell, 2002: Use of a physiological process model with forestry yield tables to set  
44 limits on annual carbon balances. *Tree Physiology*, 22(2-3), 179-188.
- 45 Warneck, P., 1988: Chemistry of the natural atmosphere. In: *International Geophysics Series*. Vol 41.  
46 [Dmowska, R. and J.R. Holton (eds.)]. Academic Press, London, 757 pp.
- 47 Warner, J., and S. Twomey, 1967: The production of cloud nuclei by cane fires and the effect on cloud  
48 droplet concentration. *Journal of Atmospheric Science*, 24, 704-706.
- 49 Warwick, N.J., S. Bekki, K.S. Law, E.G. Nisbet, and J.A. Pyle, 2002: The impact of meteorology on the  
50 interannual growth rate of atmospheric methane. *Geophysical Research Letters*, 29(20), 1947,  
51 doi:10.1029/2002GL015282.
- 52 Watson, A.J., C.S. Law, K.A. Vanscoy, F.J. Millero, W. Yao, G.E. Friedrich, M.I. Liddicoat, R.H.  
53 Wanninkhof, R.T. Barber, and K.H. Coale, 1994: Minimal effect of iron fertilization on sea-surface  
54 carbon-dioxide concentrations. *Nature*, 371, 143-145.
- 55 Wauben, W.M.F., J.P.F. Fortuin, P.F.G. van Velthoven, and H.M. Kelder, 1998: Comparison of modeled  
56 ozone distributions with sonde and satellite observations. *Journal of Geophysical Research*, 103, 3511-  
57 3530.

- 1 Weaver, C.J., P. Ginoux, N.C. Hsu, M.D. Chou, and J. Joiner, 2002: Radiative forcing of Saharan dust:  
2 GOCART model simulations compared with ERBE data. *Journal of Aerosol Science*, 59, 736-747.
- 3 Weaver, C.P., S.B. Roy, and R. Avissar, 2002: Sensitivity of simulated mesoscale atmospheric circulations  
4 resulting from landscape heterogeneity to aspects of model configuration. *Journal of Geophysical*  
5 *Research*, 107(D20), 8041, doi:10.1029/2001JD000376.
- 6 Weiss, R.F., 1974: Carbon dioxide in water and seawater: the solubility of a non-ideal gas. *Marine*  
7 *Chemistry*, 2, 203-215.
- 8 Wen, J., Z. Su, and Y. Ma, 2003: Determination of land surface temperature and soil moisture from tropical  
9 rainfall measuring mission/microwave imager remote sensing data. *Journal of Geophysical Research*,  
10 108(D2), doi:10.1029/2002JD002176.
- 11 Werdell, P.J., and C.S. Roesler, 2003: Remote assessment of benthic substrate composition in shallow  
12 waters using multispectral reflectance. *Limnology and Oceanography*, 48(1), 557-567 Part 2.
- 13 Werner, M., I. Tegen, S.P. Harrison, K.E. Kohfeld, I.C. Prentice, H. Rodhe, and C. Roelandt, 2002: Seasonal  
14 and interannual variability of the mineral dust cycle under present and glacial climate conditions.  
15 *Journal of Geophysical Research*, 107(D24), 4744, doi:10.1029/2002JD002365.
- 16 Wetzel, M.A., and L.L. Stowe, 1999: Satellite-observed patterns in stratus microphysics, aerosol optical  
17 thickness, and shortwave radiative forcing. *Journal of Geophysical Research*, 104(D24), 31287-31299.
- 18 Wetzel, P., A. Winguth, and E. Maier-Reimer, 2005: Sea-to-air CO<sub>2</sub> flux from 1948 to 2003: a model study.  
19 *Global Biogeochemical Cycles*, 19(2), GB2005, doi:10.1029/2004GB002391.
- 20 Wetzel, P., E. Maier-Reimer, M. Botzet, J. Jungclaus, N. Keenlyside, and M. Latif, Effects of ocean biology  
21 on the penetrative radiation in a coupled climate model. *Journal of Climate*, (under revision).
- 22 Whalen, R.T., and W.S. Reeburgh, 1990: Consumption of atmospheric methane by tundra soils. *Nature*, 346,  
23 160-162.
- 24 Wild, M., A. Ohmura, H. Gilgen, and D. Rosenfeld, 2004: On the consistency of trends in radiation and  
25 temperature records and implications for the global hydrological cycle. *Geophysical Research Letters*,  
26 31, doi:10.1029/2003GL019188.
- 27 Wild, M., H. Gilgen, A. Rösch, A. Ohmura, C.N. Long, E.G. Dutton, B. Forgan, A. Kallis, V. Russak, and  
28 A. Tsvetkov, 2005: From dimming to brightening: decadal changes in solar radiation at earth's surface.  
29 *Science*, 308, 847-850.
- 30 Wild, O., M.J. Prather, and H. Akimoto, 2001: Indirect long-term global radiative cooling from NO<sub>x</sub>  
31 emissions. *Geophysical Research Letters*, 28(9), 1719-1722.
- 32 Wild, O., P. Pochanart, and H. Akimoto, 2004: Trans-Eurasian transport of ozone and its precursors. *Journal*  
33 *of Geophysical Research*, 109, D11302, doi:10.1029/2003JD004501.
- 34 Williams, A.A.J., D.J. Karoly, and N. Tapper, 2001: The sensitivity of Australian fire danger to climate  
35 change. *Climatic Change*, 49, 171-191.
- 36 Williams, K.D., A. Jones, D.L. Roberts, C.A. Senior, and M.J. Woodage, 2001: The response of the climate  
37 system to the indirect effects of anthropogenic sulphate aerosols. *Climate Dynamics*, 17, 845-856.
- 38 Williamson, D., J. Boyle, R. Cederwall, M. Fiorino, J. Hnilo, J. Olson, T. Phillips, G. Potter, and S.C. Xie,  
39 2005: Moisture and temperature at the atmospheric radiation measurement southern great plains site in  
40 forecasts with the Community Atmosphere Model (CAM2). *Journal of Geophysical Research*, 110,  
41 doi:10.1029/2004JD005109.
- 42 Wong, S., W.-C. Wang, I.S.A. Isaksen, T.K. Berntsen, and J.K. Sundet, 2004: A global climate-chemistry  
43 model study of present-day tropospheric chemistry and radiative forcing from changes in tropospheric  
44 O<sub>3</sub> since the preindustrial period. *Journal of Geophysical Research*, 109, D11309,  
45 doi:10.1029/2003JD003998.
- 46 World Meteorological Organization, 2003: *Scientific assessment of ozone depletion: 2002*. Global Ozone  
47 Research and Monitoring Project - Report No. 47, Geneva, 498 pp.
- 48 Wu, W., and R.E. Dickinson, 2004: Time scales of layered soil moisture memory in the context of land-  
49 atmosphere interaction. *Journal of Climate*, 17(14), 2752-2764.
- 50 Wu, W., M.A. Geller, and R.E. Dickinson, 2002a: A case study for land model evaluation: simulation of soil  
51 moisture amplitude damping and phase shift. *Journal of Geophysical Research*, 107(D24), 4793,  
52 doi:10.1029/2001JD001405.
- 53 Wu, W., M.A. Geller, and R.E. Dickinson, 2002b: Soil moisture profile variability in response to long-term  
54 precipitation. *Journal of Hydrometeorology*, 3, 604-613.
- 55 Wuebbles, D.J., and K. Hayhoe, 2002: Atmospheric methane and global change. *Earth-Science Reviews*, 57,  
56 177-210.

- 1 Xu, Z., X. Zheng, Y. Wang, S. Han, Y. Huang, J. Zhu, and K. Butterbach-Bahl, 2004: Effects of elevated  
2 CO<sub>2</sub> and N fertilization on CH<sub>4</sub> emissions from paddy rice fields. *Global Biogeochemical Cycles*, 18,  
3 GB3009, doi:10.1029/2004GB002233.
- 4 Yang, K., T. Koike, H. Fujii, T. Tamura, X.D. Xu, L.G. Bian, and M.Y. Zhou, 2004: The daytime evolution  
5 of the atmospheric boundary layer and convection over the Tibetan Plateau: observations and  
6 simulations. *Journal of the Meteorological Society of Japan*, 82(6), 1777-1792.
- 7 Yu, H., S.C. Liu, and R.E. Dickinson, 2002: Radiative effects of aerosols on the evolution of the atmospheric  
8 boundary layer. *Journal of Geophysical Research*, 107(D12), 4142, doi:10.1029/2001JD000754.
- 9 Yurganov, L.N., T. Blumenstock, E.I. Grechko, F. Hase, E.J. Hyer, E.S. Kasischke, M. Koike, Y. Kondo, I.  
10 Kramer, F.Y. Leung, E. Mahieu, J. Mellqvist, J. Notholt, P.C. Novelli, C.P. Rinsland, H.E. Scheel, A.  
11 Schulz, A. Strandberg, R. Sussmann, H. Tanimoto, V. Velazco, R. Zander, and Y. Zhao, 2004: A  
12 quantitative assessment of the 1998 carbon monoxide emission anomaly in the Northern Hemisphere  
13 based on total column and surface concentration measurements. *Journal of Geophysical Research*,  
14 109(D15), D15305, doi:10.1029/2004JD004559.
- 15 Zavaleta, E.S., B.D. Thomas, N.R. Chiariello, G.P. Asner, M.R. Shaw, and C.B. Field, 2003a: Plants reverse  
16 warming effect on ecosystem water balance. *Proceedings of the National Academy of Sciences of the*  
17 *USA*, 100(17), 9892-9893.
- 18 Zavaleta, E.S., M.R. Shaw, N.R. Chiariello, B.D. Thomas, E.E. Cleland, C.B. Field, and H.A. Mooney,  
19 2003b: Grassland responses to three years of elevated temperature, CO<sub>2</sub>, precipitation, and N  
20 deposition. *Ecological Monographs*, 73(4), 585-604.
- 21 Zeebe, R.E., and D. Archer, 2005: Feasibility of ocean fertilisation and its impact on future atmospheric CO<sub>2</sub>  
22 levels. *Geophysical Research Letters*, 32, L09703, doi:10.1029/2005GL022449.
- 23 Zeebe, R.E., and D. Wolf-Gladrow, 2001: CO<sub>2</sub> in seawater: equilibrium, kinetics, isotopes. *Elsevier*  
24 *Oceanography Series*, 65, Elsevier, Amsterdam, 346 pp.
- 25 Zehr, J.P., and B.B. Ward, 2002: Nitrogen cycling in the ocean: new perspectives on processes and  
26 paradigms. *Applied and Environmental Microbiology*, 68(3), 1015-1024.
- 27 Zender, C., R.L. Miller, and I. Tegen, 2004: Quantifying mineral dust mass budgets: systematic terminology,  
28 constraints, and current estimates. *EOS*, 85(48).
- 29 Zeng, G., and J.A. Pyle, 2003: Changes in tropospheric ozone between 2000 and 2100 modeled in a  
30 chemistry-climate model. *Geophysical Research Letters*, 30, 1392, doi:10.1029/2002GL016708.
- 31 Zeng, N., K. Hales, and J.D. Neelin, 2002: Nonlinear dynamics in a coupled vegetation-atmosphere system  
32 and implications for desert-forest gradient. *Journal of Climate*, 15, 3474-3485.
- 33 Zeng, N., H. Qian, E. Munoz, and R. Iacono, 2004: How strong is carbon cycle-climate feedback under  
34 global warming? *Geophysical Research Letters*, 31, L20203, doi:10.1029/2004GL020904.
- 35 Zeng, X., M. Shaikh, Y. Dai, and R.E. Dickinson, 2002: Coupling of the common land model to the NCAR  
36 community climate model. *Journal of Climate*, 15, 1832-1854.
- 37 Zeng, X.-D., S.S.P. Shen, X. Zeng, and R.E. Dickinson, 2004: Multiple equilibrium states and the abrupt  
38 transitions in a dynamical system of soil water interacting with vegetation. *Geophysical Research*  
39 *Letters*, 31, L05501, doi:10.1029/2003GL018910.
- 40 Zhang, H., J.L. McGregor, A. Henderson-Sellers, and J.J. Katzfey, 2004: Impacts of land surface model  
41 complexity on a regional simulation of a tropical synoptic event. *Journal of Hydrometeorology*, 5, 180-  
42 198.
- 43 Zhang, J., U. Lohmann, and P. Stier, 2005: A microphysical parameterization for convective clouds in the  
44 ECHAM5 climate model, 1. Single column results evaluated at the Oklahoma ARM site. *Journal of*  
45 *Geophysical Research*, 110, doi:10.1029/2004JD005128.
- 46 Zhang, X.Y., R. Arimoto, and Z.S. An, 1997: Dust emission from Chinese desert sources linked to variations  
47 in atmospheric circulation. *Journal of Geophysical Research*, 102, 28041-28047.
- 48 Zhang, X.Y., H.Y. Lu, R. Arimoto, and S.L. Gong, 2002: Atmospheric dust loadings and their relationship to  
49 rapid oscillations of the Asian winter monsoon climate: two 250-kyr loess records. *Earth and Planetary*  
50 *Science Letters*, 202, 637-643.
- 51 Zhang, X.Y., S.L. Gong, T.L. Zhao, R. Arimoto, Y.Q. Wang, and Z.J. Zhou, 2003: Sources of Asian dust  
52 and role of climate change versus desertification in Asian dust emission. *Geophysical Research Letters*,  
53 30, doi:10.1029/2003GL018206, 2272.
- 54 Zhang, Y.-C., W.B. Rossow, A.A. Lacis, V. Oinas, and M.I. Mishchenko, 2004: Calculation of radiative flux  
55 profiles from the surface to top-of-atmosphere based on ISCCP and other global data sets: refinements  
56 of the radiative transfer model and the input data. *Journal of Geophysical Research*, 109, D19105, doi:  
57 10.1029/2003JD004457

- 1 Zhou, L., C.J. Tucker, R.K. Kaufmann, D. Slayback, N.V. Shabanov, and R.B. Myneni, 2001: Variations in  
2 northern vegetation activity inferred from satellite data of vegetation index during 1981 to 1999.  
3 *Journal of Geophysical Research*, 106, 20069-20083.
- 4 Zhou, L., R.E. Dickinson, Y. Tian, M. Jin, K. Ogawa, H. Yu, and T. Schmugge, 2003a: A sensitivity study  
5 of climate and energy balance simulations with use of satellite-derived emissivity data over Northern  
6 Africa and the Arabian Peninsula. *Journal of Geophysical Research*, 108(D24), 4795, doi:  
7 so.1029/2003JD004083.
- 8 Zhou, L., R.E. Dickinson, K. Ogawa, Y. Tian, M. Jin, T. Schmugge, and E. Tsvetsinskaya, 2003b: Relations  
9 between albedos and emissivities from MODIS and ASTER data over North African desert.  
10 *Geophysical Research Letters*, 30(20), 2026, doi:10.1029/2003GL018069.
- 11 Zhou, L., R.K. Kaufmann, Y. Tian, R.B. Myneni, and C.J. Tucker, 2003c: Relation between interannual  
12 variations in satellite measures of northern forest greenness and climate between 1982 and 1999.  
13 *Journal of Geophysical Research*, 108(D1), 4004, doi:10.1029/2002JD002510.
- 14 Zhou, L., R.E. Dickinson, Y. Tian, X. Zeng, Y. Dai, Z.-L. Yang, C.B. Schaaf, F. Gao, Y. Jin, A. Strahler,  
15 R.B. Myneni, H. Yu, W. Wu, and M. Shaikh, 2003d: Comparison of seasonal and spatial variations of  
16 albedos from Moderate-Resolution Imaging Spectroradiometer (MODIS) and common land model. *Journal*  
17 *of Geophysical Research*, 108(D15), 4488, doi:10.1029/2002JD003326.
- 18 Zhou, L., R.E. Dickinson, Y. Tian, J. Fang, Q. Li, R.K. Kaufmann, C.J. Tucker, and R.B. Myneni, 2004:  
19 Evidence for a significant urbanization effect on climate in China. *Proceedings of the National*  
20 *Academy of Sciences USA*, 101(26), 9540-9544.
- 21 Zhuang, Q., J.M. Melillo, D.W. Kicklinghter, R.G. Prinn, A.D. McGuire, P.A. Steudler, B.S. Felzer, and S.  
22 Hu, 2004: Methane fluxes between terrestrial ecosystem and the atmosphere at northern high latitudes  
23 during the past century: a retrospective analysis with a process-based biogeochemistry model. *Global*  
24 *Biogeochem.Cycles*, 18, doi:10.1029/2008GB002239.
- 25 Ziemke, J.R., and S. Chandra, 1999: Seasonal and interannual variabilities in tropical tropospheric ozone.  
26 *Journal of Geophysical Research*, 104(21), 425–21, 442.
- 27 Zittel, W., and M. Altmann, 1996: Molecular hydrogen and water vapour emissions in a global hydrogen  
28 energy economy. Proceedings of the 11<sup>th</sup> World Hydrogen Energy Conference, Stuttgart, Germany,  
29 June 1996. Schön & Wetzel, Frankfurt am Main, Germany, pp. 71-82.
- 30 Zondervan, I., R.E. Zeebe, B. Rost, and U. Riebesell, 2001: Decreasing marine biogenic calcification: a  
31 negative feedback on rising  $p\text{CO}_2$ . *Global Biogeochemical Cycles*, 15, 507-516.
- 32 Zu, Z., X. Zheng, Y. Wang, S. Han, Y. Huang, J. Zhu, and K. Butterbach-Bahl, 2004: Effects of elevated  
33  $\text{CO}_2$  and N fertilization of  $\text{CH}_4$  emissions from paddy rice fields. *Global Biogeochemical Cycles*, 18,  
34 GB3009, doi:10.1029/2004GB002233.
- 35  
36

1 **Tables**2  
3 **Table 7.3.3.** Couplings between climate and ocean biogeochemical processes.  
4

Process	Primary Forcings	Direction	Quantitative Potential	Timescale (years)	Certainty	Important Associated Impacts and Secondary Feedbacks
Sea water buffering	Atmospheric $p\text{CO}_2$ increase	-	high	0–1000	certainty	Dominant inorganic feedback, leads to significant ocean acidification
Changes in $\text{CO}_2$ solubility, dissociation, and buffer factor	Warming, atmospheric $p\text{CO}_2$ increase, slowing of ocean circulation	+	medium		certainty	pH lowering with impact on biology and sediments
Biological export production of POC, DOC storage	Warming, $p\text{CO}_2$ increase, runoff loads, dust deposition, slowing of ocean circulation, change in radiation	unknown	unknown	0–1000	indication	Complex feedback chain, involving food chain and biodiversity, $\text{N}_2\text{O}$ production, light absorption and heating, particle dynamics
Biological export production of PIC ( $\text{CaCO}_3$ )	$p\text{CO}_2$ increase (pH decrease)	unknown	unknown	0–1000	indication	Ballast effect and particle dynamics, DMS production, species shifts
Coral growth	Warming, atmospheric $p\text{CO}_2$ increase (pH decrease)	-	unknown	0–100	certainty	Albedo change, biodiversity, species shifts
Dissolution of $\text{CaCO}_3$ sediments	$p\text{CO}_2$ increase (pH decrease)	-	high	1000–100000	certainty	biodiversity
DMS production	$\text{CaCO}_3$ production, dust flux	unknown	unknown	0–100	indication	tropospheric cloud formation
Destabilization of gas hydrates	warming	+	unknown	unknown	potential	Bacterial blooms
Purposeful $\text{CO}_2$ storage	Human intention to mitigate climate change	unknown	unknown	unknown	potential	Ecological impact on water column and bottom fauna, may increase fossil fuel burning

5  
6

**Table 7.4.5.** Global budgets of tropospheric ozone ( $\text{Tg yr}^{-1}$ ) for the present-day atmosphere<sup>a</sup>

Reference	Model	Stratosphere-Troposphere Exchange (STE)	Chemical Production <sup>b</sup>	Chemical Loss <sup>b</sup>	Dry Deposition	Burden (Tg)	Lifetime <sup>c</sup> (days)
TAR <sup>d</sup>	11 models	770 ± 400	3420 ± 770	3470 ± 520	770 ± 180	300 ± 30	24 ± 2
Lelieveld and Dentener (2000)	TM3	570	3310	3170	710	350	33
Bey et al. (2001) <sup>e</sup>	GEOS-Chem	470	4900	4300	1070	320	22
Sudo et al. (2002)	CHASER	593	4895	4498	990	322	25
Horowitz et al. (2003)	MOZART-2	340	5260	4750	860	360	23
Von Kuhlmann et al. (2003)	MATCH-MPIC	540	4560	4290	820	290	21
Shindell et al. (2003)	GISS	417	NR <sup>f</sup>	NR	1470	349	NR
Hauglustaine et al. (2004)	LMDz-INCA	523	4486	3918	1090	296	28
Park et al. (2004)	UMD-CTM	480	NR	NR	1290	340	NR
Rotman et al. (2004)	IMPACT	660	NR	NR	830	NR	NR
Wong et al. (2004)	SUNYA/UiO GCCM	600	NR	NR	1100	376	NR
Stevenson et al. (2004)	STOCHEM	395	4980	4420	950	273	19
Wild et al. (2004)	FRSGC/UCI	520	4090	3850	760	283	22
Stevenson et al. (2005)	25 models	520 ± 200	5060 ± 570	4560 ± 720	1010 ± 220	340 ± 40	22 ± 2

Notes:

(a) From global model simulations describing the atmosphere of the last decade of the 20th century.

(b) Chemical production and loss rates are calculated for the odd oxygen family, usually defined as  $\text{O}_x = \text{O}_3 + \text{O} + \text{NO}_2 + 2\text{NO}_3 + 3\text{N}_2\text{O}_5 + \text{HNO}_4 + \text{peroxyacylnitrates}$  (and sometimes  $\text{HNO}_3$ ), to avoid accounting for rapid cycling of ozone with short-lived species that have little implication for its budget. Chemical production is mainly contributed by reactions of NO with peroxy radicals, while chemical loss is mainly contributed by the  $\text{O}(^1\text{D}) + \text{H}_2\text{O}$  reaction and by the reactions of ozone with  $\text{HO}_2$ , OH, and alkenes. Several models in this table do not report production and loss separately ("NR" entry in the table), reporting instead net production. However, net production is not a useful quantity for budget purposes because (1) it represents a small residual between large production and loss, (2) it represents the balance between STE and dry deposition, both of which are usually parameterized as a flux boundary condition.

(c) Calculated as the ratio of the burden to the sum of chemical and deposition losses

(d) Means and standard deviations from an ensemble of 11 global model budgets from the 1996–2000 literature reported in the TAR. The mean budget does not balance exactly because only 9 CTMs reported their chemical production and loss statistics.

(e) A more recent version of GEOS-Chem by Martin et al. (2003b) gives identical rates and burdens.

(f) Not reported

---

## Chapter 7: Couplings Between Changes in the Climate System and Biogeochemistry

**Coordinating Lead Authors:** Guy Brasseur, Kenneth Denman

**Lead Authors:** Amnat Chidthaisong, Philippe Ciais, Peter Cox, Robert Dickinson, Didier Hauglustaine, Christoph Heinze, Elisabeth Holland, Daniel Jacob, Ulrike Lohmann, Srikanthan Ramachandran, Pedro Leite da Silva Diaz, Steven Wofsy, Xiaoye Zhang

**Contributing Authors:** David Archer, V. Arora, John Austin, D. Baker, Joe Berry, Gordon Bonan, Philippe Bousquet, Deborah Clark, V. Eyring, Johann Feichter, Pierre Friedlingstein, Inez Fung, Sandro Fuzzi, Sunling Gong, Alex Guenther, Ann Henderson-Sellers, Andy Jones, Bernd Kärcher, Mikio Kawamiya, Yadvinder Malhi, K. Masarie, Surabi Menon, J. Miller, P. Peylin, A. Pitman, Johannes Quaas, P. Rayner, Ulf Riebesell, C. Rödenbeck, Leon Rotstayn, Nigel Roulet, Chris Sabine, Martin Schultz, Michael Schulz, Will Steffen, Steve Schwartz, J. Lee-Taylor, Yuhong Tian, Oliver Wild, Liming Zhou.

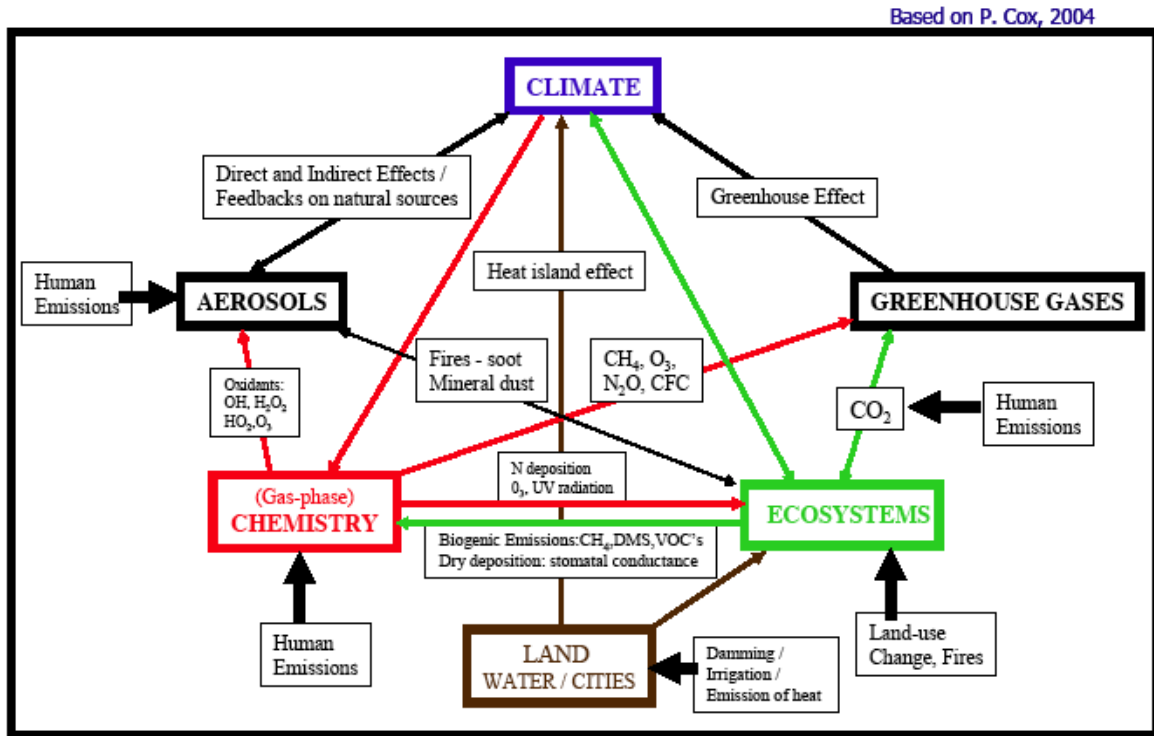
**Review Editors:** Kansri Boonpragob, Martin Heimann, Mario Molina

**Date of Draft:** 12 August 2005

**Notes:** This is the TSU compiled version

---

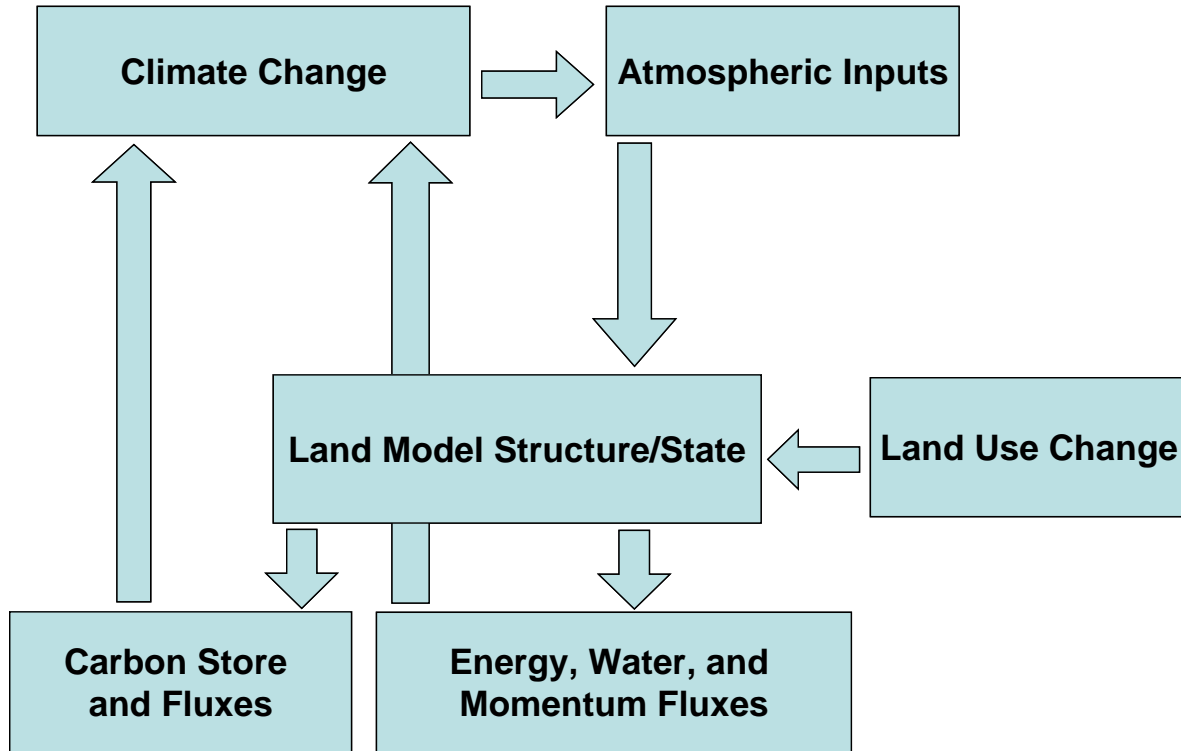
1  
2  
3  
4



5  
6  
7  
8  
9  
10  
11

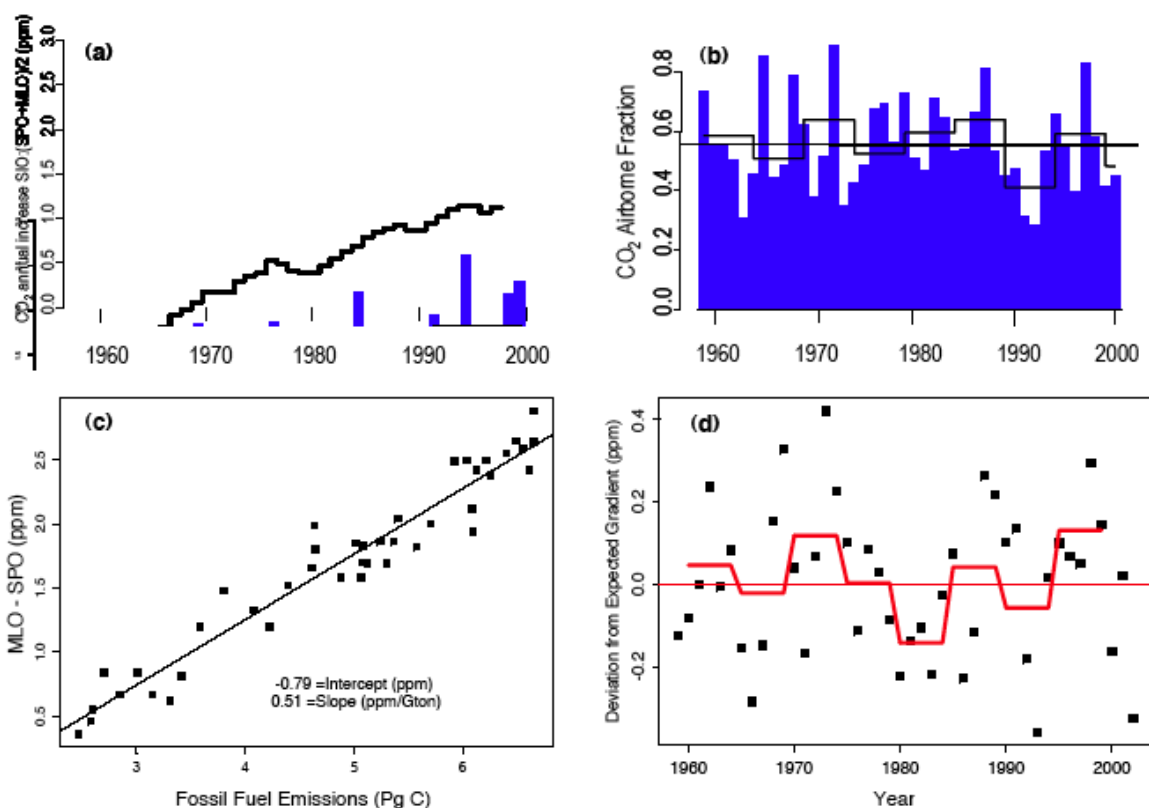
**Figure 7.1.1.** Schematic representation of some key interactions in the Earth system between climate, greenhouse gases, chemically reactive gases, aerosols and ecosystems. The effects of human activities on different elements of the Earth system are indicated.

1  
2



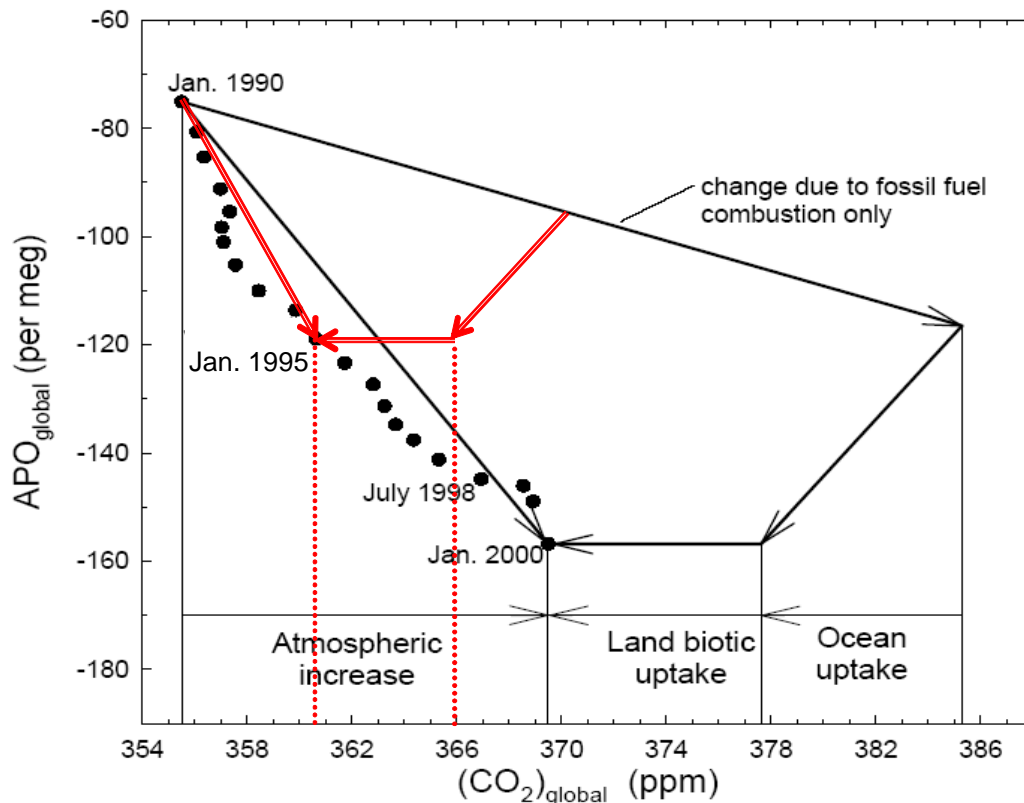
3  
4  
5  
6  
7  
8  
9

**Figure 7.2.1.** A schematic depiction of the various factors that must be addressed in evaluating the role of land in anthropogenic climate change.

1  
23  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13  
14  
15  
16  
17

**Figure 7.3.1.** CO<sub>2</sub> emissions and concentrations in the atmosphere. (a) Annual increases in global CO<sub>2</sub> concentrations as indicated by the mean of Mauna Loa (MLO) and South Pole (SPO) annual averages (*blue bars*), and 5-yr block averages of these data, from Keeling and Whorf (2004; SIO data, *black line*) also shown are data from Conway and Tans (2004, CMDL data, 5-yr block averages (1978–2003), *red line*). The upper stepped *bold black line* shows CO<sub>2</sub> annual increases if 100% of fossil fuel emissions stayed in the atmosphere. (b) Fraction of fossil fuel emissions remaining in the atmosphere each year (“Airborne fraction”, *blue bars*), 5-yr mean (*stepped line —*) (SIO data), and mean since 1958 (*broken line ---*). Note the anomalously low airborne fraction in the early 1990s. (c) North-south concentration difference, as indicated by MLO — SPO (ppm), plotted against annual fossil fuel emission flux (Pg-C), 1959–2003. (d) Deviation of the annual mean concentration different (MLO – SPO) from the line in panel (c), with 5-yr block averages in *red —*.

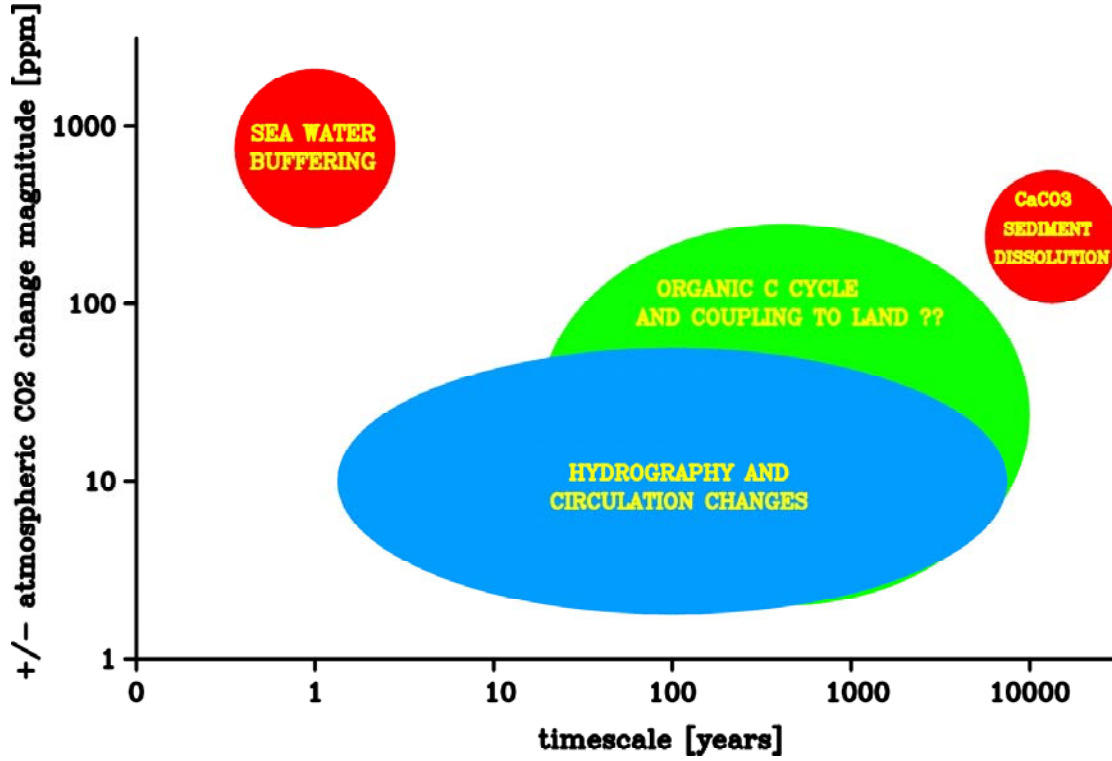
1  
2



3  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13

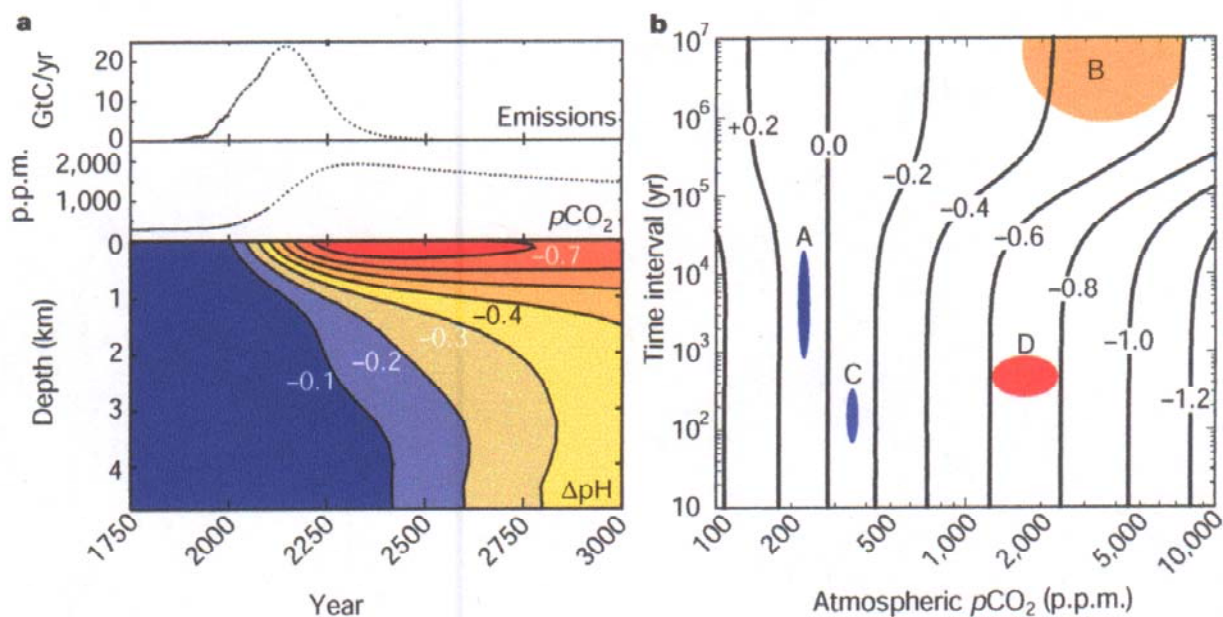
**Figure 7.3.2.** Semi-annual changes in Atmospheric Potential Oxygen (APO):  $\Delta APO = \Delta O_2 + \alpha_B \Delta CO_2$ , where  $\alpha_B$  represents the average  $O_2:CO_2$  molar exchange ratio ( $\cong 1.10$ ) for land biotic photosynthesis and respiration and  $\Delta O_2$  and  $\Delta CO_2$  are defined from global data for the  $O_2:N_2$  ratio and for  $CO_2$  mole fraction, respectively. APO is defined so that uptake or release by land biota has no effect. Fossil fuel inputs lower APO (slope  $(\alpha_B - \alpha_\phi)$  due to the higher value of  $\alpha_\phi$  ( $\cong 1.4$ ). Dissolution of  $CO_2$  in the ocean has slope  $\alpha_B$  since  $O_2$  is not taken up. The analysis corrected for  $\Delta O_2$  associated with outgassing of  $O_2$  associated with warming of the upper ocean. From Manning and Keeling (2005).

1  
2



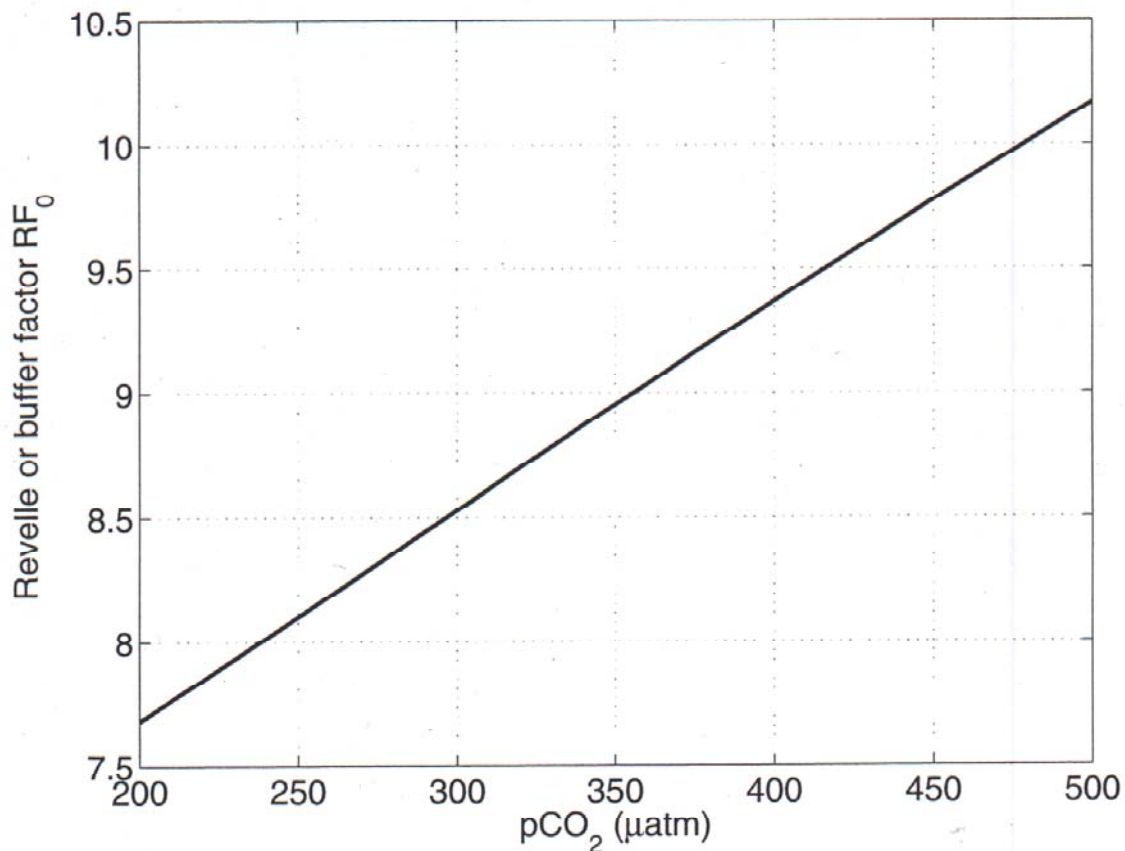
3  
4  
5  
6  
7

Figure 7.3.3. Overview of biogeochemical ocean climate feedbacks (see also Table 7.3.2.2.1).

1  
23  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13  
14

**Figure 7.3.4.** Atmospheric release of CO<sub>2</sub> from the burning of fossil fuels may give rise to a marked increase in ocean acidity. (a, left) Atmospheric CO<sub>2</sub> emissions, historical atmospheric CO<sub>2</sub> levels and predicted CO<sub>2</sub> concentrations from this emissions scenario, together with changes in ocean pH based on horizontally averaged chemistry. (b, right) Estimated maximum change in surface ocean pH as a function of final atmospheric CO<sub>2</sub> pressure, and the transition time over which this CO<sub>2</sub> pressure is linearly approached from 280 μatm. A. Glacial-interglacial changes; B. slow changes over the past 300 Myr; C. historical changes in ocean surface waters; D. unabated fossil-fuel burning over the next few centuries. Figure and caption from Caldeira and Wickett (2003).

1



2

3

4

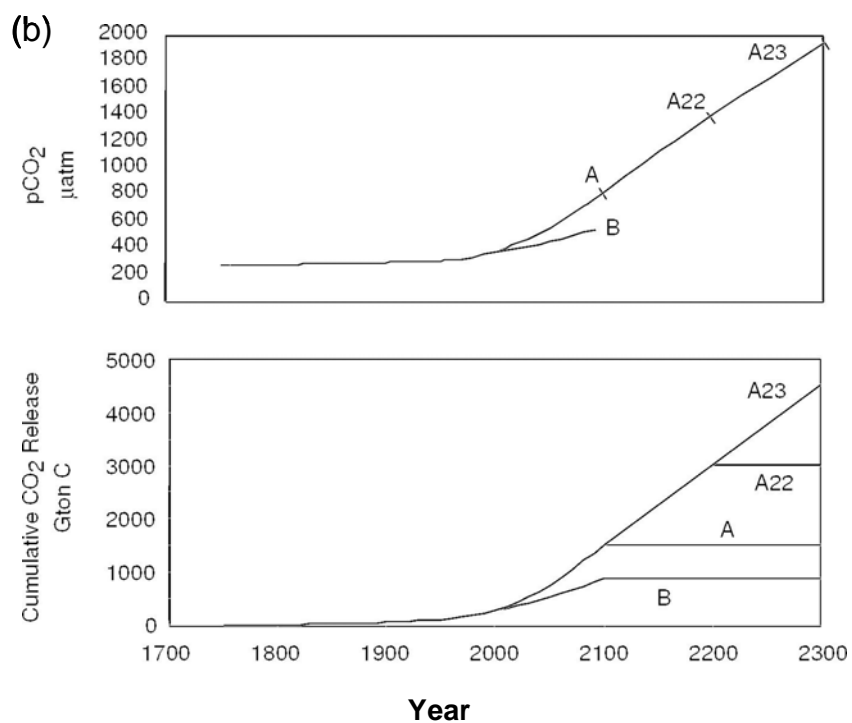
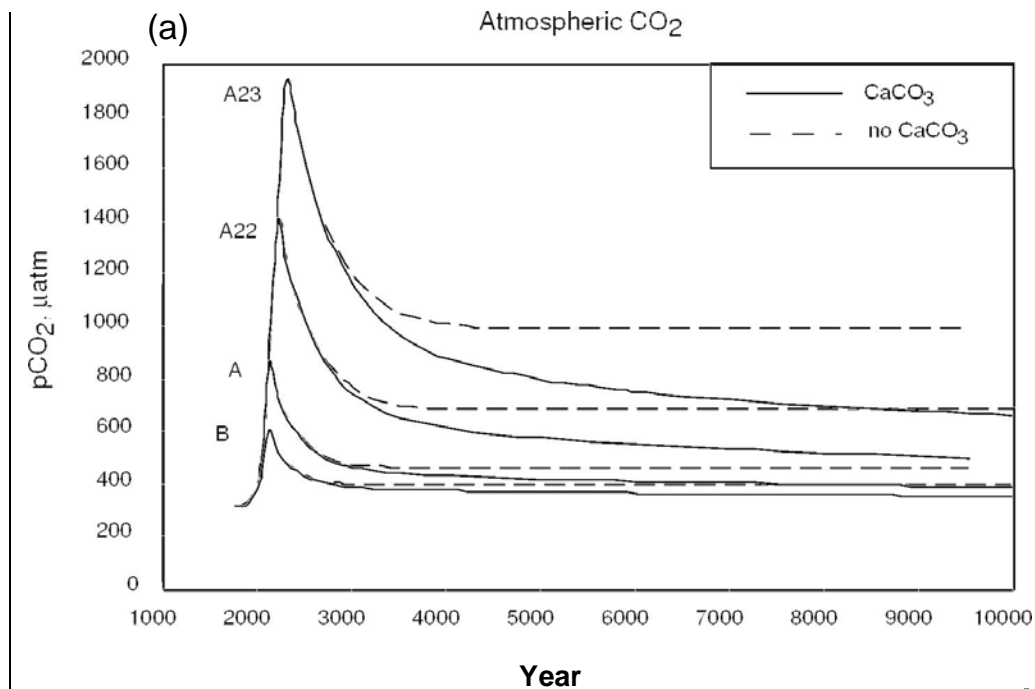
**Figure 7.3.5.** The Revelle factor  $RF_0$  as a function of  $pCO_2$  ( $T_C = 25^\circ$ ,  $S = 35$ ,  $TA = 2300 \mu\text{mol kg}^{-1}$ ). With increasing partial pressure of  $CO_2$  the Revelle factor increases and thus the buffering capacity of the ocean decreases. Figure and caption from Zeebe and Wolf-Gladrow (2001).

6

7

8

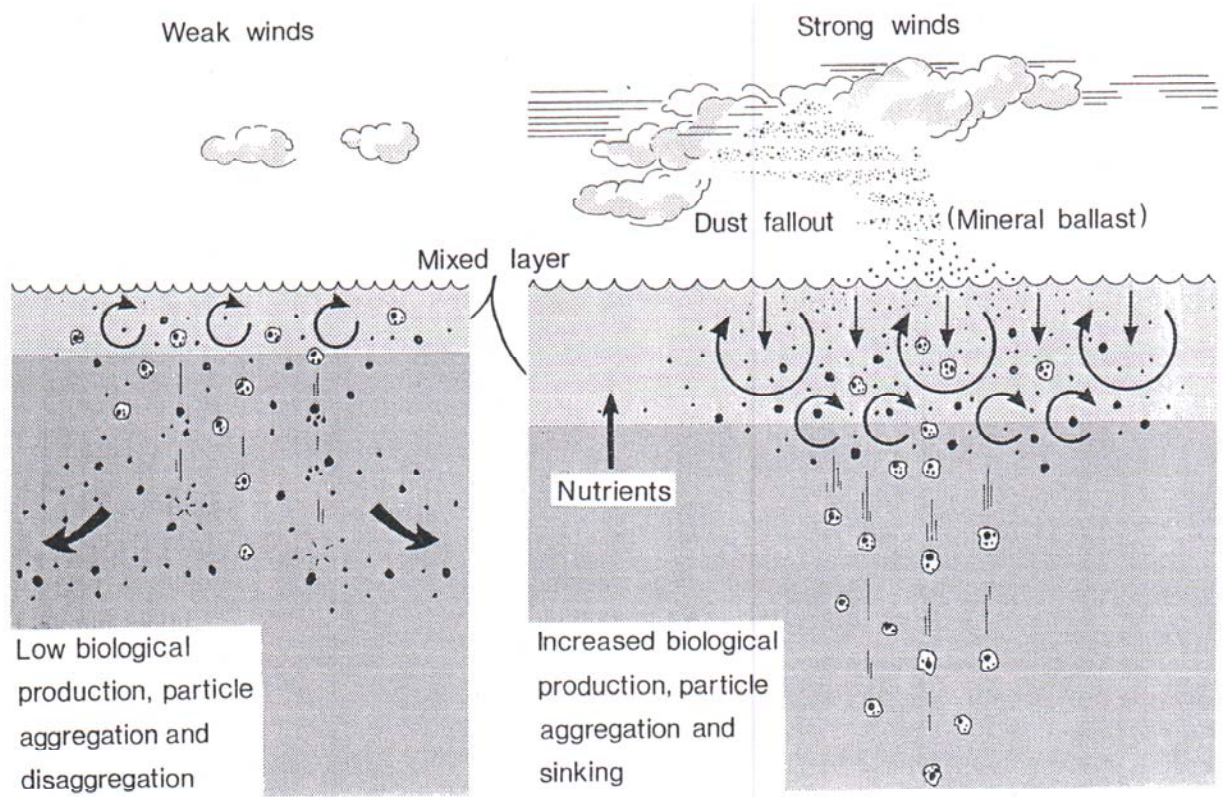
1  
2



3  
4  
5  
6  
7  
8  
9  
10  
11

**Figure 7.3.6.** Panel a: Model response to anthropogenic CO<sub>2</sub> release to the year AD 10,000, including the effect of CaCO<sub>3</sub> dissolution from sediments (solid curve), and neglecting CaCO<sub>3</sub> (dashed curve). The pCO<sub>2</sub> is held to IPCC projections for scenarios A and B to the year 2100 and extrapolated to the years 2200 and 2300 at the scenario A year 2100 emission rate for runs B, A, A22, and A23 respectively. After those times, a zero net terrestrial release of CO<sub>2</sub> is specified. Panel b: Emission scenarios used. (From Archer et al., 1998)

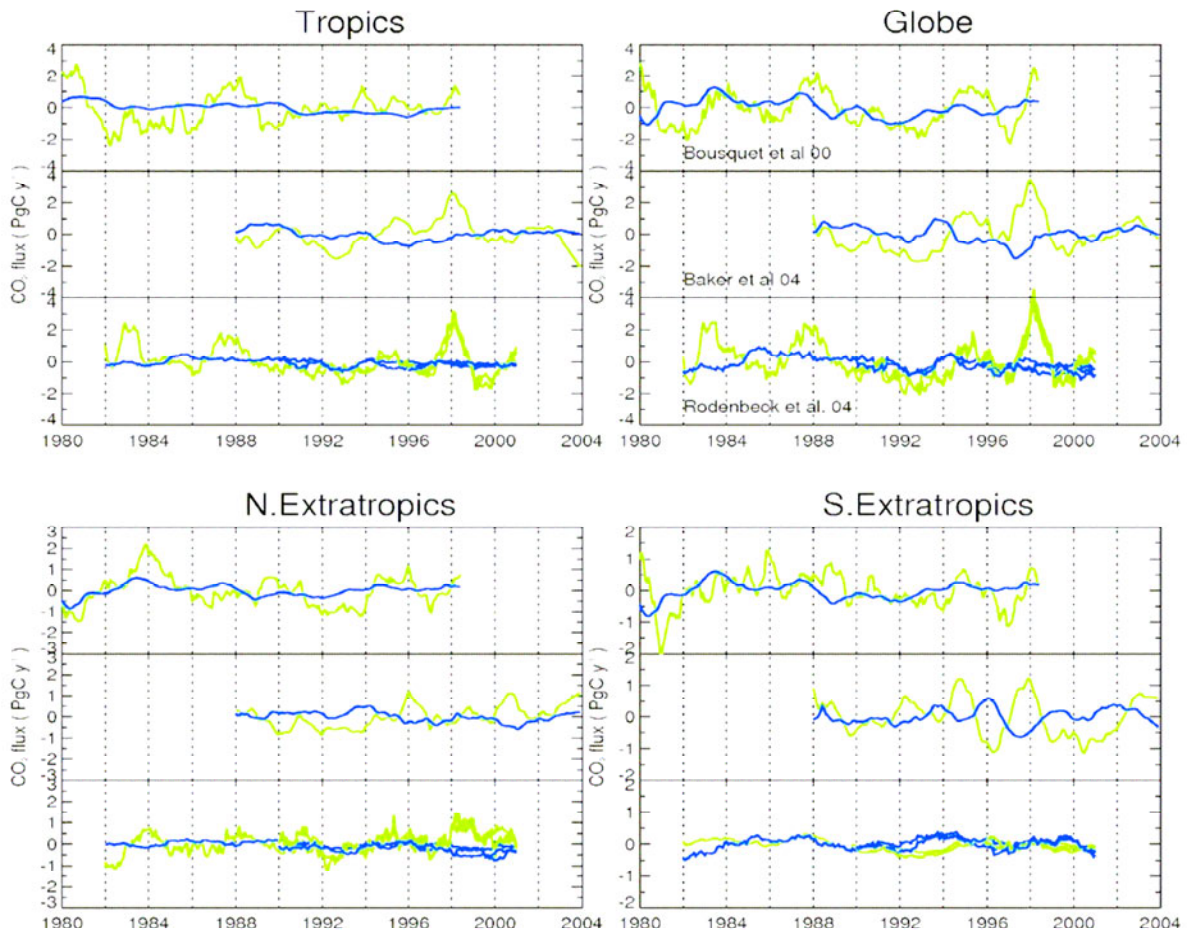
1  
2



3  
4  
5  
6  
7  
8

**Figure 7.3.7.** The wind-driven biological pump and its enhancement by atmospheric dust deposition over the sea. Figure and caption from Ittekkot (1993).

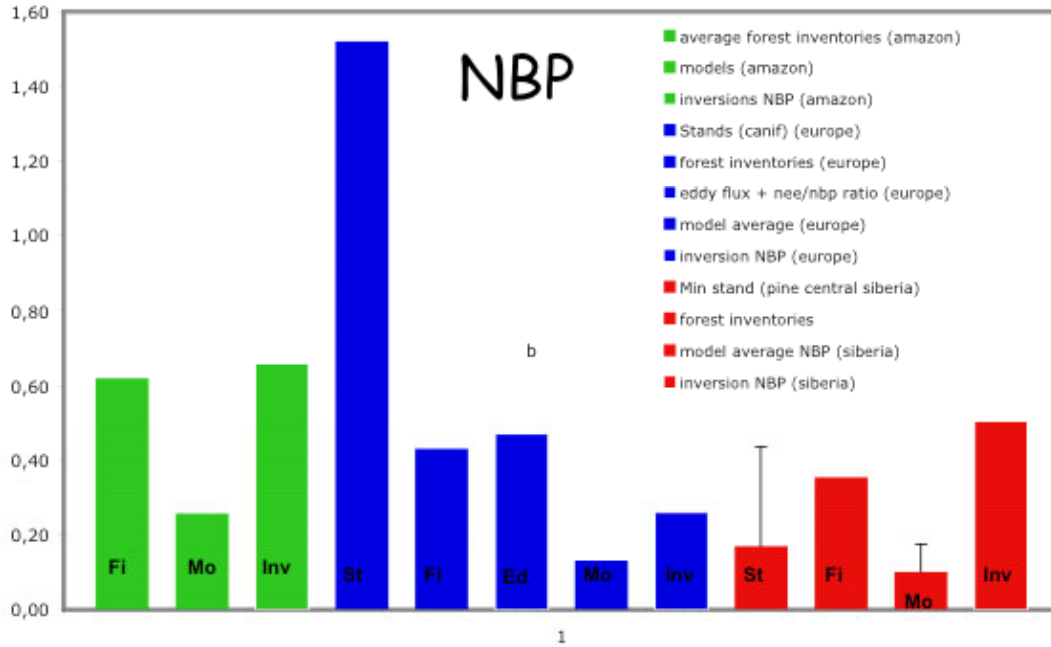
1  
2



3  
4  
5  
6  
7  
8  
9  
10

**Figure 7.3.8.** Ocean and land fluxes year-to-year variations from inversions summed into large latitude bands, and over the globe during approx. the past 20 years. Three different inversion ensembles are shown, and for each flux and each region, the long term mean values has been subtracted and the seasonal signal has been removed.

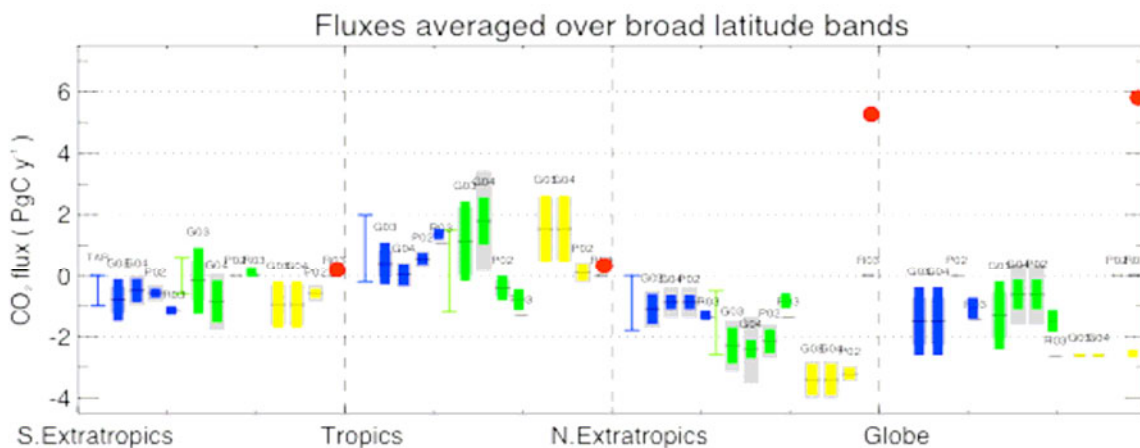
1  
2



3  
4  
5  
6  
7  
8  
9

**Figure 7.3.9.** Estimates of the mean terrestrial carbon balance (in tC per ha per year) from different approaches at various scales and for three distinct biomes, **Green** = Amazon ; **Blue** = Western Europe ; **Red** = Siberia (data compiled by Ciais et al., 2005).

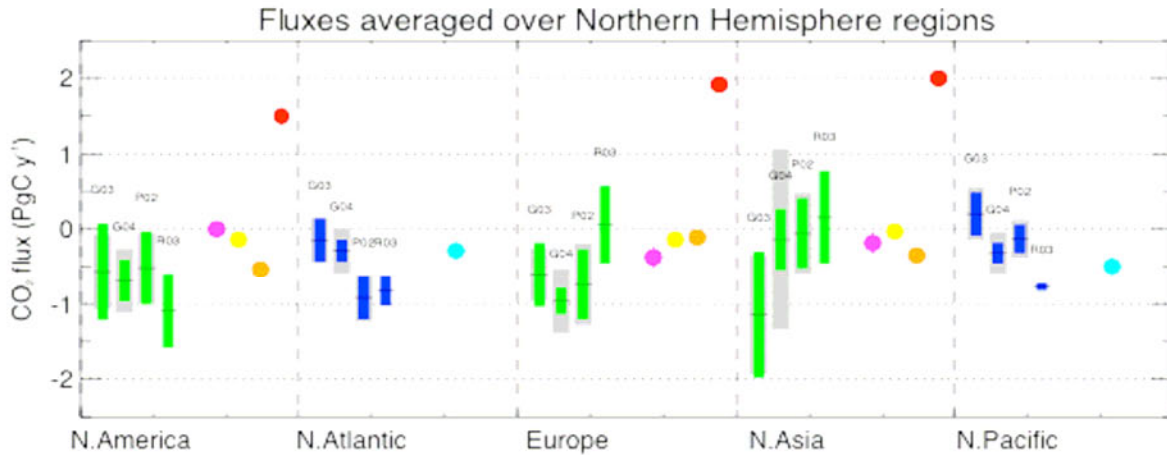
1  
2



3  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13  
14  
15  
16  
17  
18  
19

**Figure 7.3.10.** Ocean and land fluxes from inversions summed into large latitude bands, and over the globe during the period 1992–1996. **Blue** = Ocean fluxes, **Green** = land fluxes, **Yellow** = total land plus ocean fluxes obtained after subtracting the non-optimized fossil fuel emissions in **red**. The mean flux from different ensembles of inversions is reported together with random errors and the range of bias due to different settings within each ensemble. Error boxes in colour are the average of 1-s Gaussian random errors returned by each member of the ensemble. Grey error boxes are the spread of the mean fluxes from different arbitrary settings. **TAR** = range of mean fluxes from Third Assessment Report ; **G02** = ensemble using annual mean CO<sub>2</sub> observations with grey error from 16 transport models (Gurney et al., 2002); **G03** is the ensemble using smoothed monthly CO<sub>2</sub> observations with grey error from 13 transport models (also from Gurney et al., 2003) ; **P04** is the ensemble with grey error from 3 transport models times 3 grouping of large regions times 3 inversion set of regions (Peylin et al., 2005a); **R03** is the ensemble where the fluxes are solved for many small regions using monthly flask data, with the grey error from different sensitivity inversions (Rodenbeck et al., 2003).

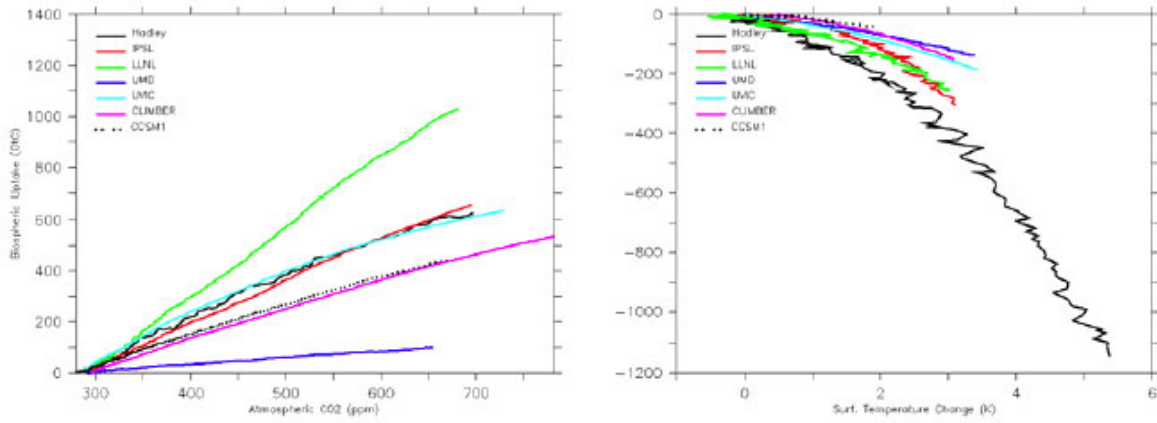
1  
2



3  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13

**Figure 7.3.11.** Blue, Green = Ocean and land fluxes from inversions in the Northern Hemisphere. Bottom-up continental-wide or ocean-basin-wide estimates are given separately. Magenta = Forest biomass inventory data for Europe (Nabuurs et al., 1997) and Siberia (Shvidenko and Nilsson, 2003); Yellow = Forest biomass inventory data including Russia, Mongolia and China (Goodale et al., 2002); Orange = bottom-up synthesis of the net carbon balance of coterminous U.S. (Pacala et al., 2001), Geographic Europe (Janssens et al., 2003), and Russia (Shvidenko and Nilsson, 2003). Cyan = air-sea basin scale fluxes (Takahashi et al., 1999).

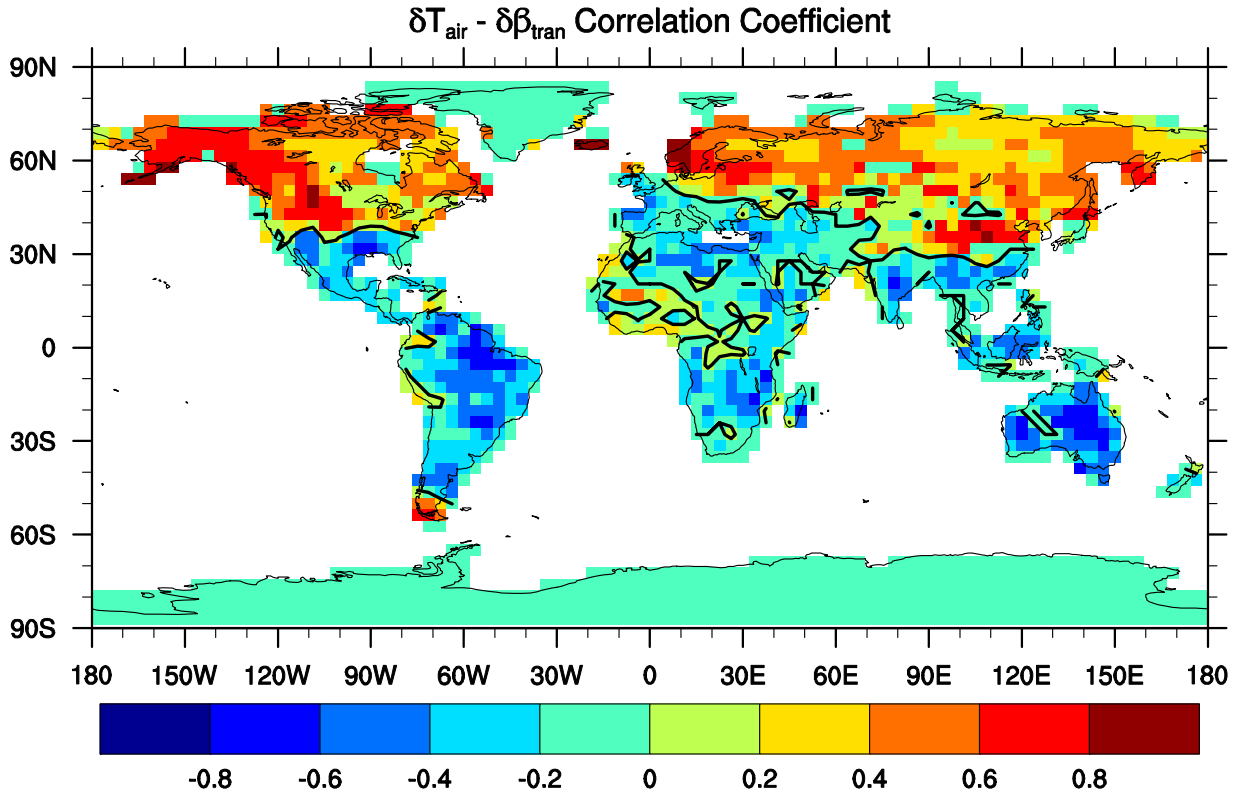
1  
2



3  
4  
5  
6  
7  
8  
9  
10

**Figure 7.3.12.** (a) Change in land carbon storage (Pg-C) versus atmospheric CO<sub>2</sub> concentration (ppm) for the C<sup>4</sup>MIP uncoupled simulations. The mean gradient of each line is the parameter  $\beta_L$ . (b) Change in land carbon storage (Pg-C) with global warming (K). In this case the mean gradient of each line is the parameter  $\gamma_L$ . (c) Like (a), but for  $\beta_O$ . (d) Like (b), but for  $\gamma_O$ .

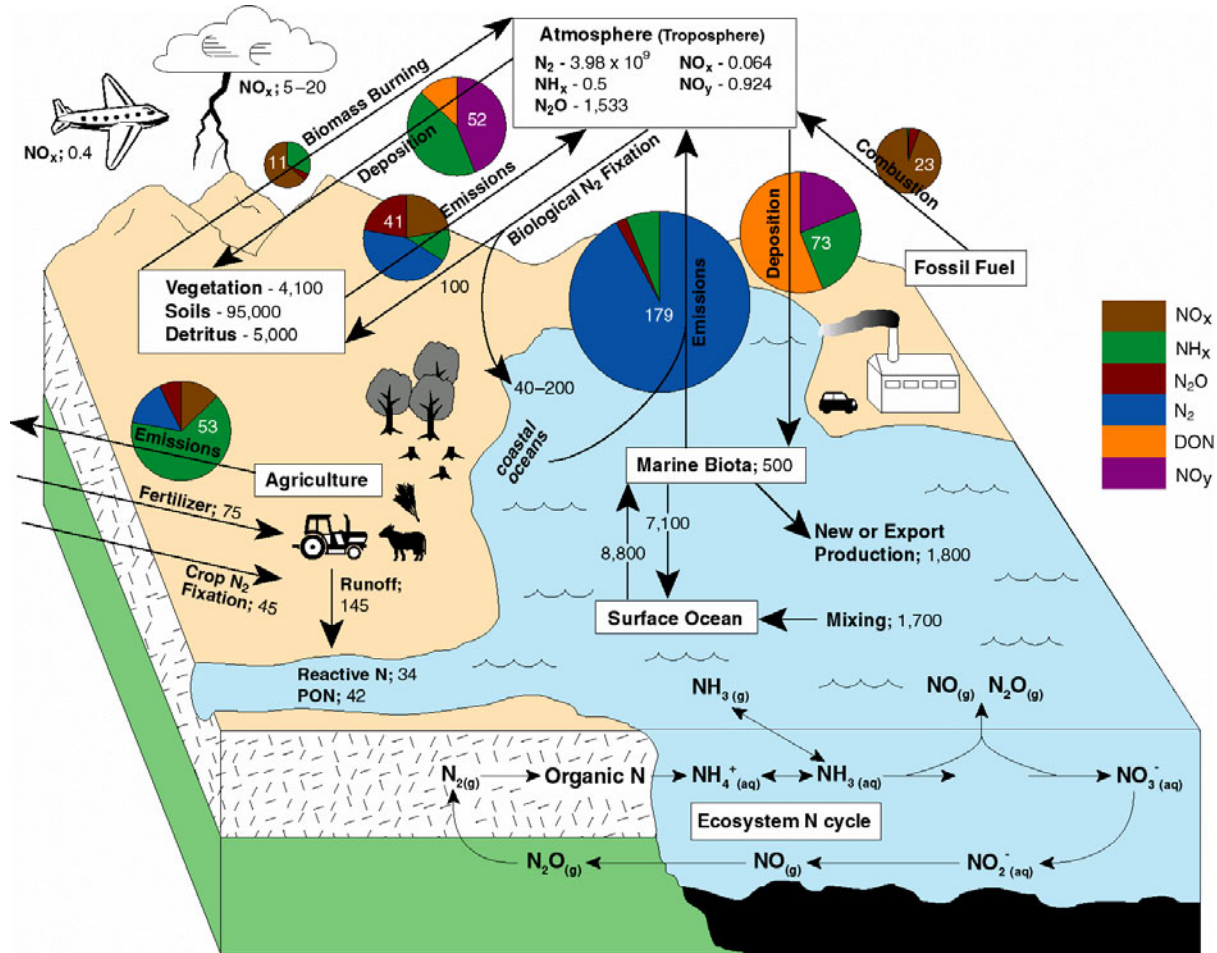
1  
2



3  
4  
5  
6  
7  
8  
9

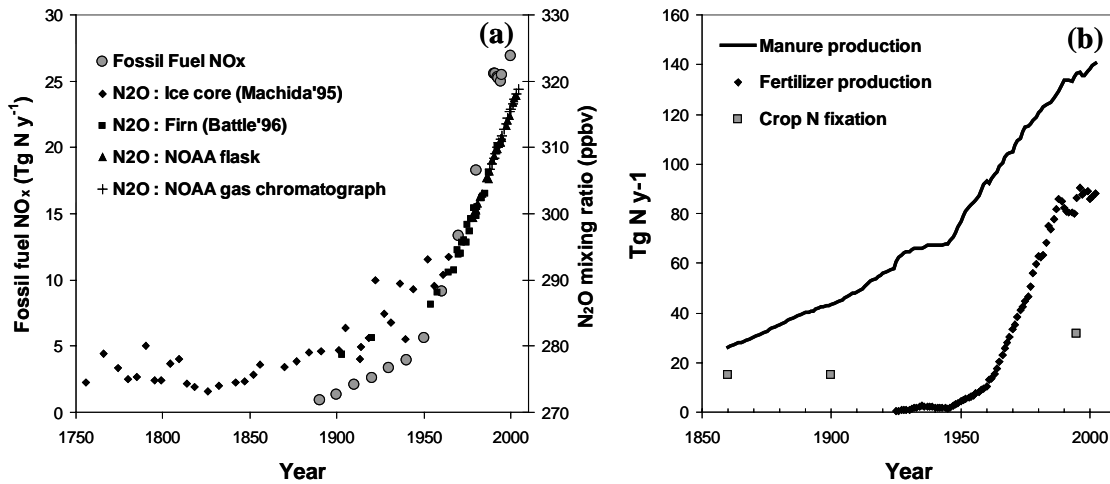
**Figure 7.3.13.** Correlations between temperature and soil moisture changes between the coupled and uncoupled runs from the NCAR-CSM1 model, showing the tendency towards warmer-drier conditions in the tropics and warmer-moister conditions at high latitudes (from Fung et al., 2005).

1  
2

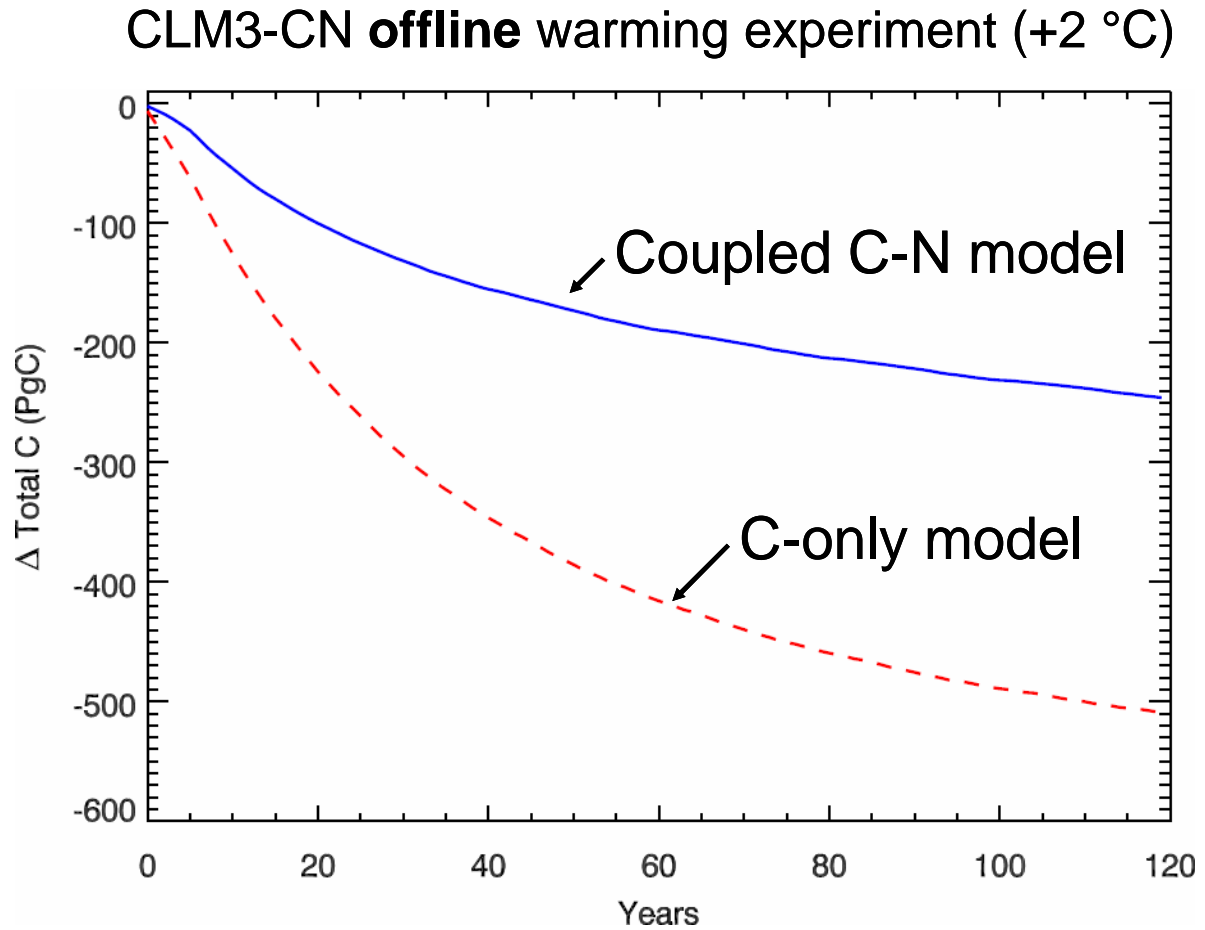


3  
4  
5  
6  
7  
8  
9  
10  
11  
12

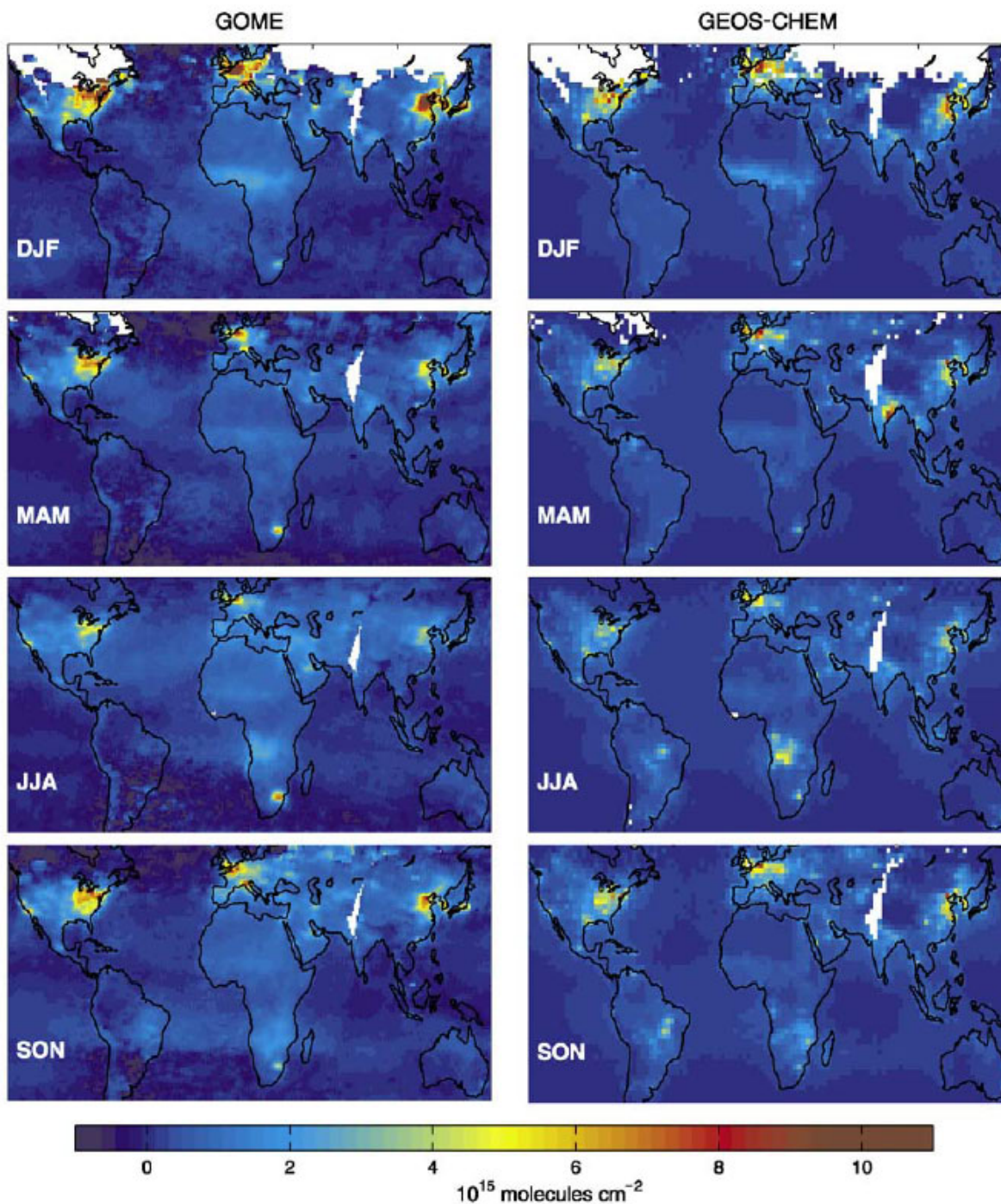
**Figure 7.4.1.** Key fluxes and components of the global nitrogen cycle, including emissions and deposition. Each of the key atmospheric species including  $\text{NO}_x$  ( $\text{NO} + \text{NO}_2$ ),  $\text{NH}_x$  ( $\text{NH}_3 + \text{NH}_4^+$ ),  $\text{N}_2\text{O}$ ,  $\text{N}_2$ , DON (Dissolved Organic Nitrogen) and  $\text{NO}_y$  (total odd nitrogen =  $\text{NO}_x + \text{HNO}_3 + \text{HONO} + \text{HO}_2\text{NO}_2 + \text{NO}_3 +$  nitrate radical + Peroxyacetyl nitrates +  $\text{N}_2\text{O}_5$  + organic nitrates) and their fluxes are shown. The ecosystem nitrogen cycle depicts the internal transformations nitrogen going starting with  $\text{N}_2$ , and the transformation, nitrification and denitrification, back to  $\text{N}_2$  to complete the full cycle. All units are in Tg of N, Tg =  $10^{12}$  g.

1  
23  
4  
5  
6  
7  
8  
9  
10  
11

**Figure 7.4.2.** (a) Changes in the emissions of fossil fuel NO<sub>x</sub> and atmospheric N<sub>2</sub>O mixing ratios since 1750. N<sub>2</sub>O mixing ratios provide the atmospheric measurement constraint on global changes to the nitrogen cycle. (b) Changes in the indices of global agricultural nitrogen cycle since 1850: the production of manure, fertilizer and estimates of crop nitrogen fixation. For data sources see <http://www-eosdis.ornl.gov/> and <http://www.cmdl.noaa.gov/>.

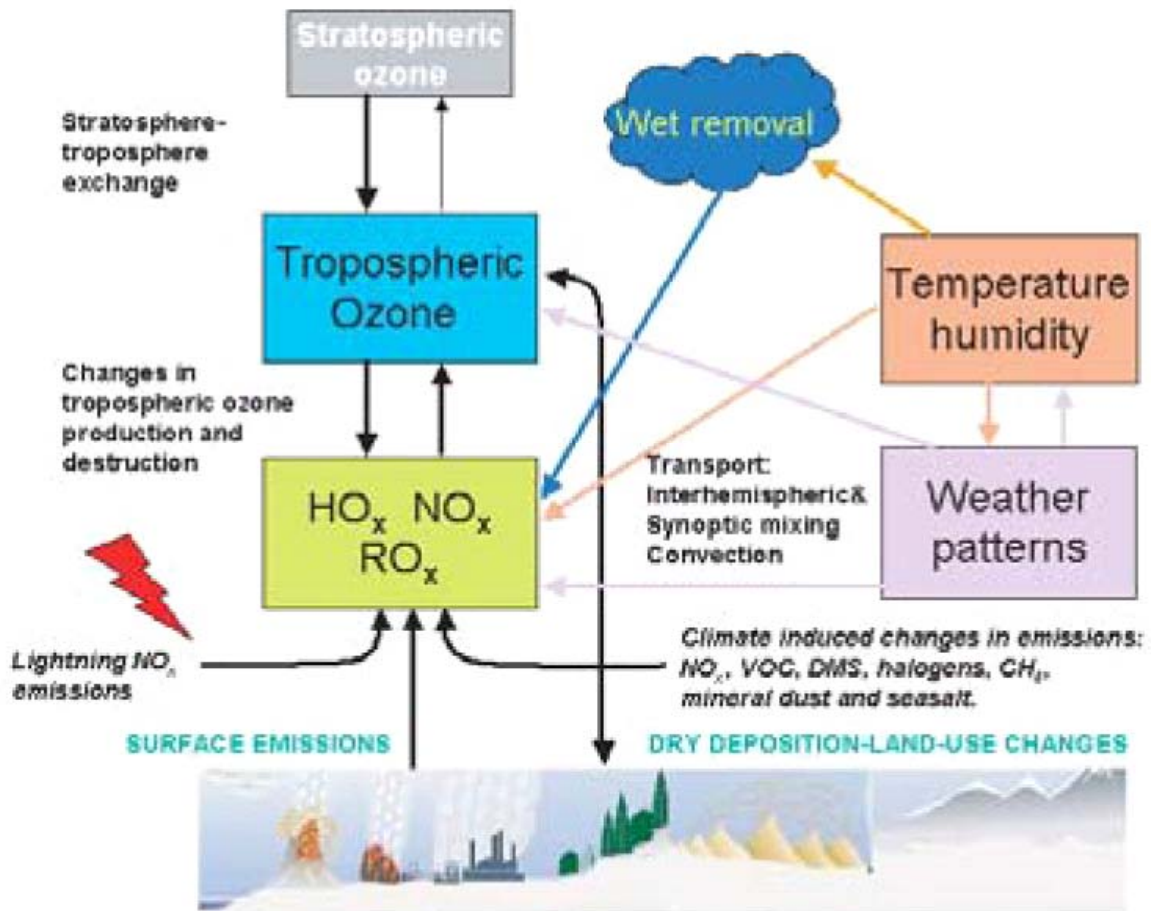
1  
23  
4  
5  
6  
7  
8  
9  
10

**Figure 7.4.3.** Simulated net global terrestrial carbon release using a carbon model and a coupled carbon nitrogen model. The simulations were done using the Community Land Model (Bonan version 3.0 modified to include carbon and carbon and nitrogen). The simulation was a step function 2°C increase in temperature forcing of the model applied after a spin-up of the carbon or carbon nitrogen model.

1  
23  
4  
5  
6  
7  
8  
9

**Figure 7.4.4.** Seasonal mean tropospheric NO<sub>2</sub> columns for September 1996–August 1997. Left: GOME retrievals. Right: GEOS-CHEM model simulation sampled along GOME overpasses and using sources from Table 7.4.6 (Bey et al., 2001). White areas have no GOME data. From Martin et al. (2003b)

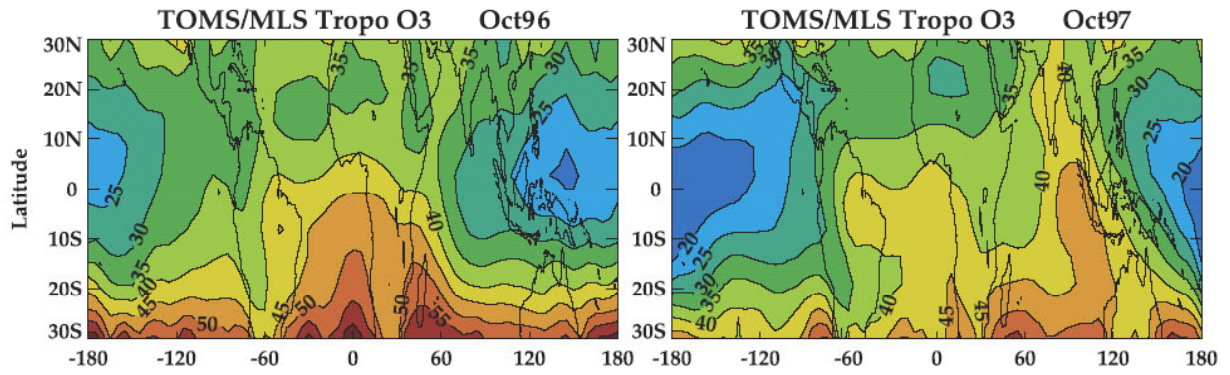
1  
2



3  
4  
5  
6  
7

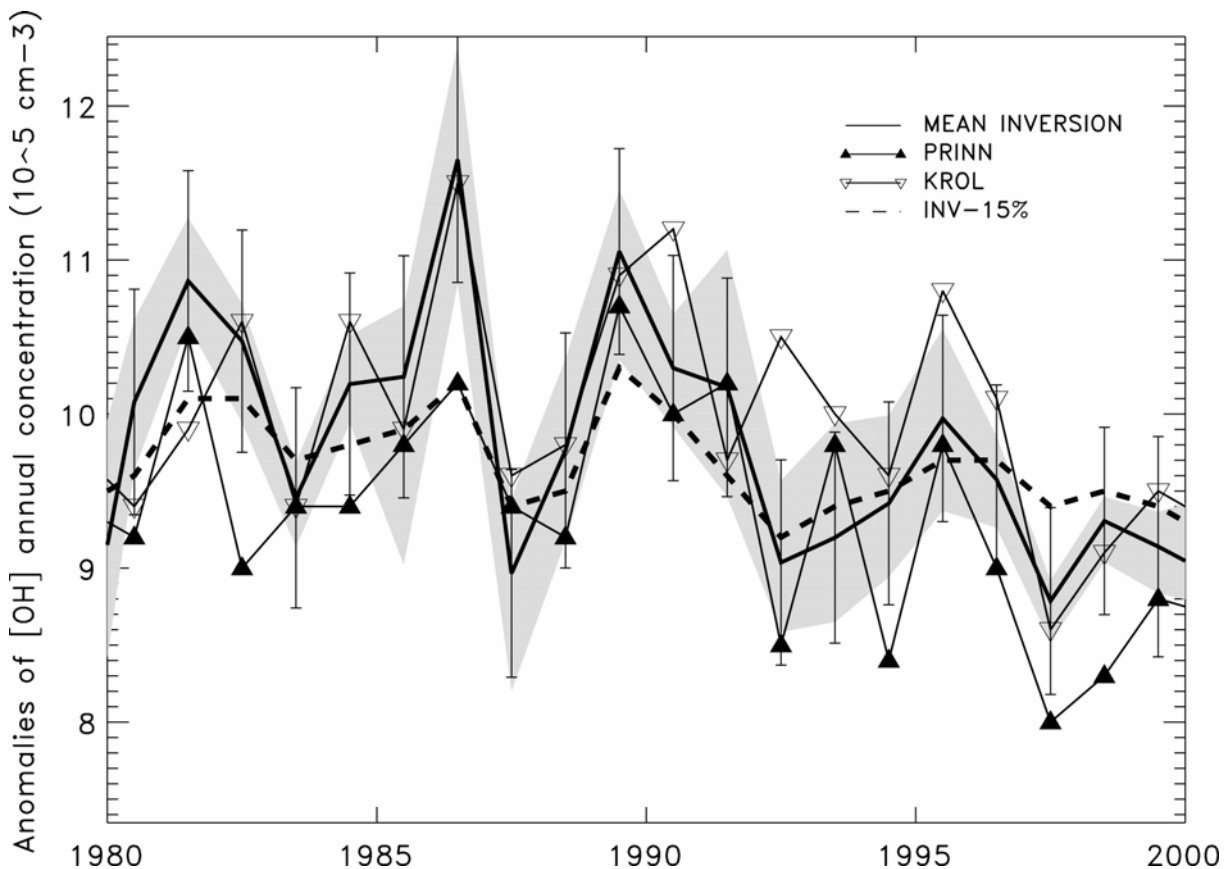
**Figure 7.4.5.** Climate variables affecting tropospheric ozone. From European Commission (2003).

1  
2



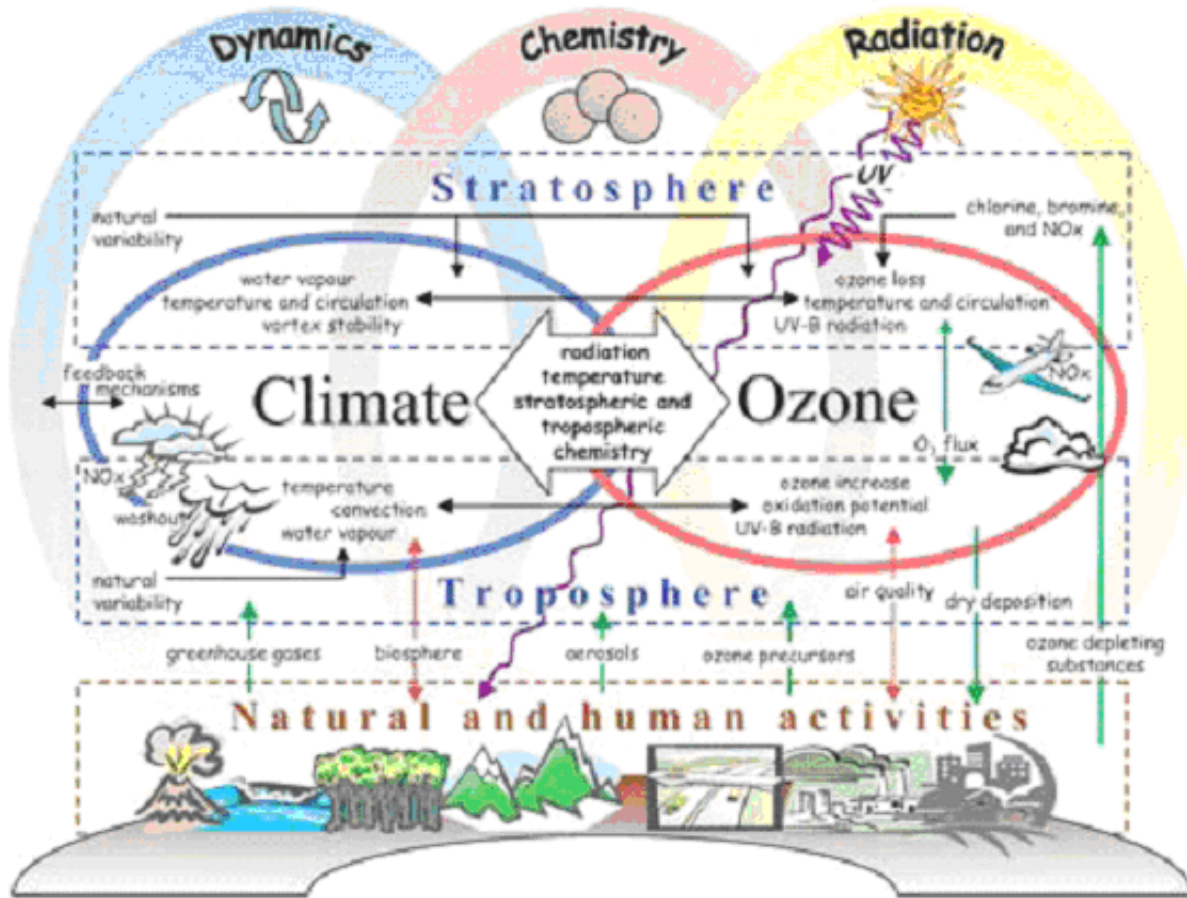
3  
4  
5  
6  
7  
8  
9

**Figure 7.4.6.** ENSO effect on tropospheric ozone columns as observed from TOMS/MLS. Monthly mean columns (DU) are shown for October 1996 (cold phase) and October 1997 (warm phase). From Chandra et al. (2003).

1  
23  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13

**Figure 7.4.7.** Annual and global mean OH variations calculated by Bousquet et al. (2005) as the mean of a set of inversions of  $\text{CH}_3\text{CCl}_3$  observations from ALE/GAGE/AGAGE network. Loose prior OH uncertainties are applied in all 16 inversions ( $\pm 100\%$ ). The mean inversion (solid line) is compared with estimates from Prinn et al. (2001) (open triangles) and from Krol et al. (2003) (filled triangles). The shaded area denotes the envelope of all inversions. Dashed line is the results of an inversion in which OH prior error is tighter ( $\pm 15\%$ ) but still gives  $\text{CH}_3\text{CCl}_3$  sources that are compatible with the inventory of McCulloch and Midgley (2001) at  $\pm 2\sigma$  level.

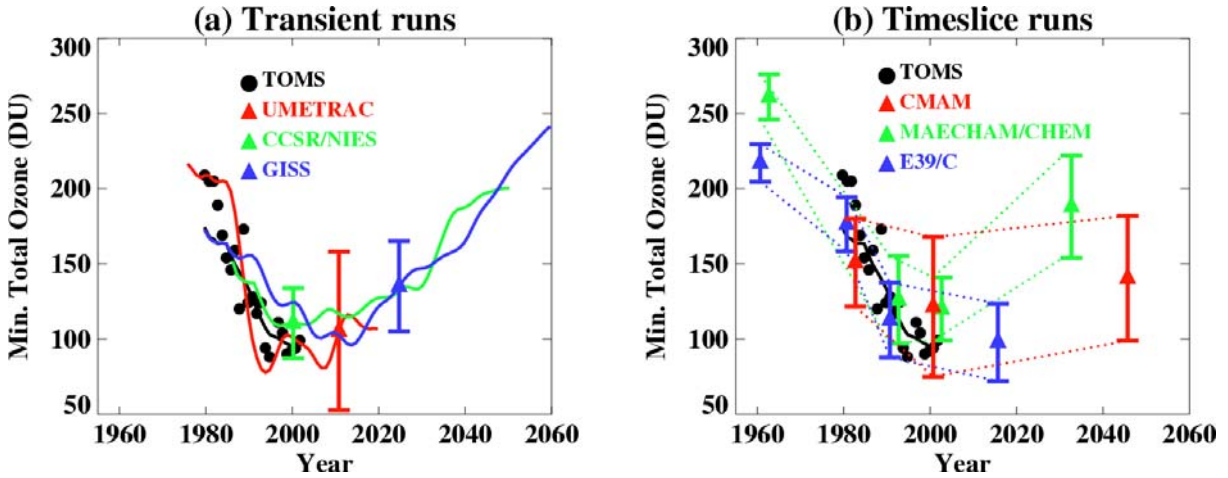
1  
2



3  
4  
5  
6  
7  
8  
9

**Figure 7.4.8.** Processes determining the ozone climate interactions in the troposphere and the stratosphere (European Commission, 2003). Atmospheric regions are indicated by blue, and source regions by brown dashed boxes.

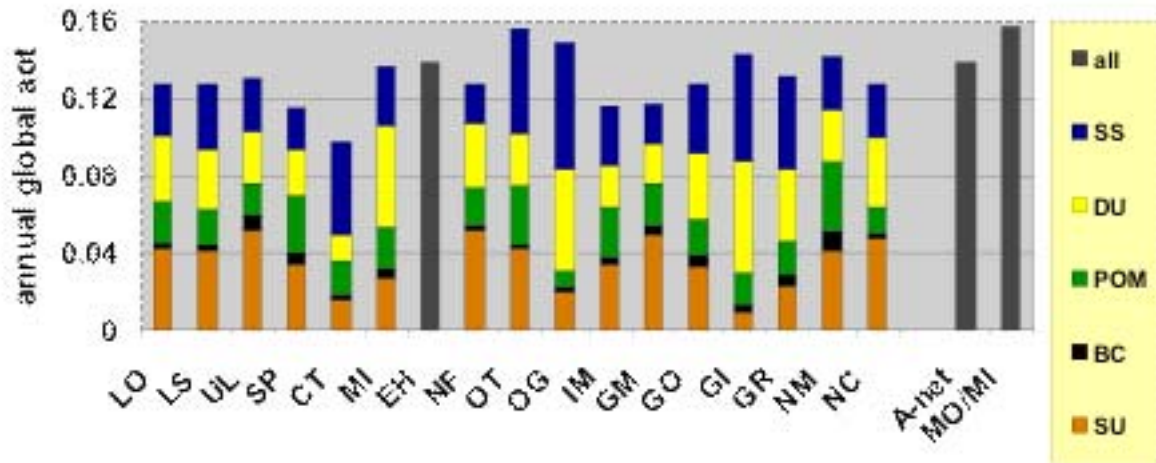
1  
2



3  
4  
5  
6  
7  
8  
9  
10  
11

**Figure 7.4.9.** Minimum Antarctic (September to November) total ozone evolution for (a) transient runs and (b) time slice runs of different CCM model experiments in comparison with TOMS data for the period 1960 to 2060. The solid lines in (a) show the results of a Gaussian smoother applied to the individual year’s results. The error bars denote twice the standard deviation of the individual years from the smoothed curve. For the time slice experiments the dotted lines are drawn between the end points of the error bars to assist in estimating trends (Austin et al., 2003; their Figure 10).

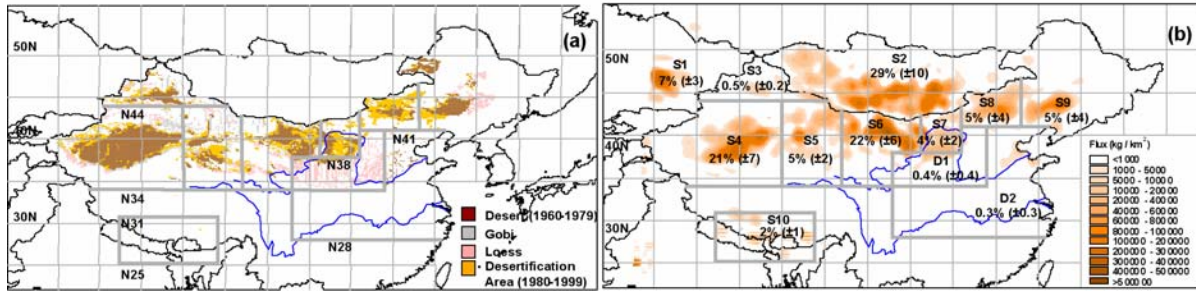
1  
2



3  
4  
5  
6  
7  
8  
9  
10  
11

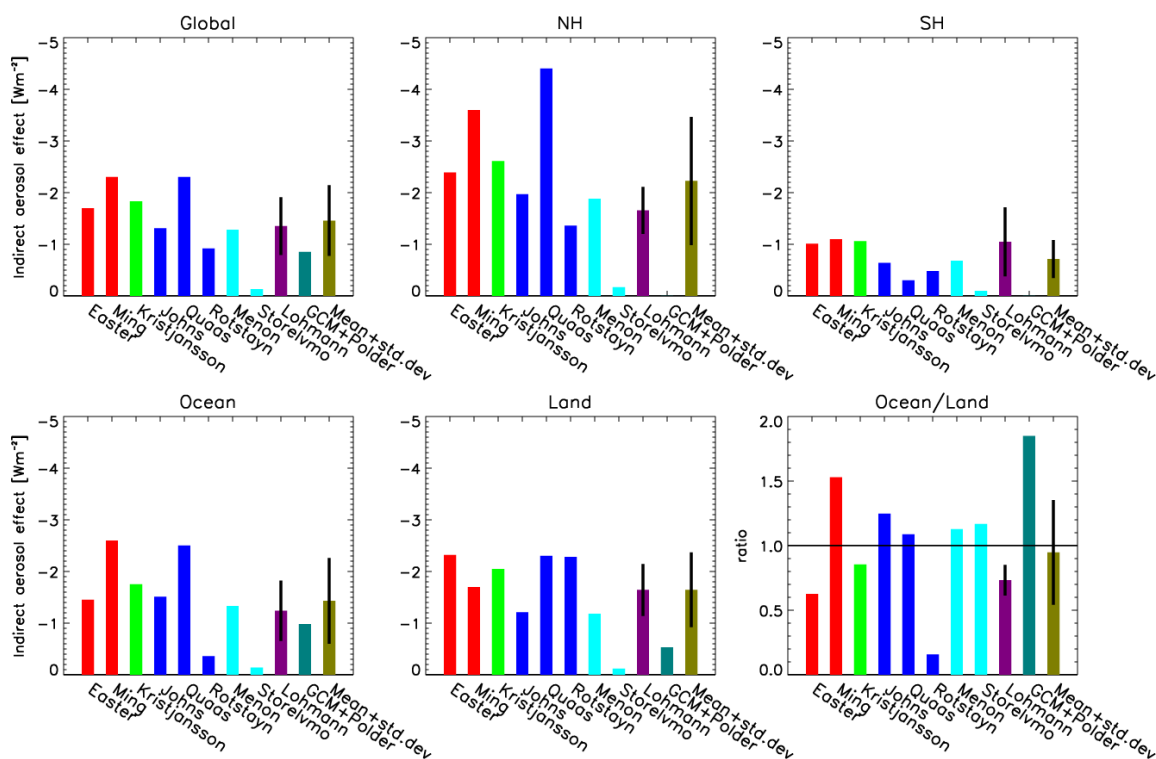
**Figure 7.5.1.** Individual contributions of the five aerosol components (SS-seasalt, DU-dust, POM-particulate organic matter, BC-black carbon, SU-sulphate) to the annual global aerosol optical thickness (at 550nm) from different global models. For comparison, two aot data references from remote sensing are provided: AERONET and a satellite-composite of MODIS (ocean) and MISR (land) data. Adapted from Kinne et al. (2005).

1  
2  
3  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13  
14



**Figure 7.5.2.** (a) Chinese desert distributions from 1960–1979 and desert plus desertification areas from 1980–1999; (b) Sources (S1 to S10) and typical depositional areas (D1 and D2) for Asian dust indicated by spring average dust emission flux ( $\text{kg km}^{-2} \text{ months}^{-1}$ ) between 1960–2002. The percentages with standard deviation in the parenthesis denote the average amount of dust production in each source region and the total amount of emissions between 1960–2002. The deserts in Mongolia (S2) and in western (S4) and northern (S6) China (mainly the Taklimakan and Badain Juran, respectively) can be considered as the major sources for Asian dust emissions. Several areas with more expansions of deserts (S7, S8, S9 and S5) are not key sources. Adapted from Zhang et al., (2003).

1  
2

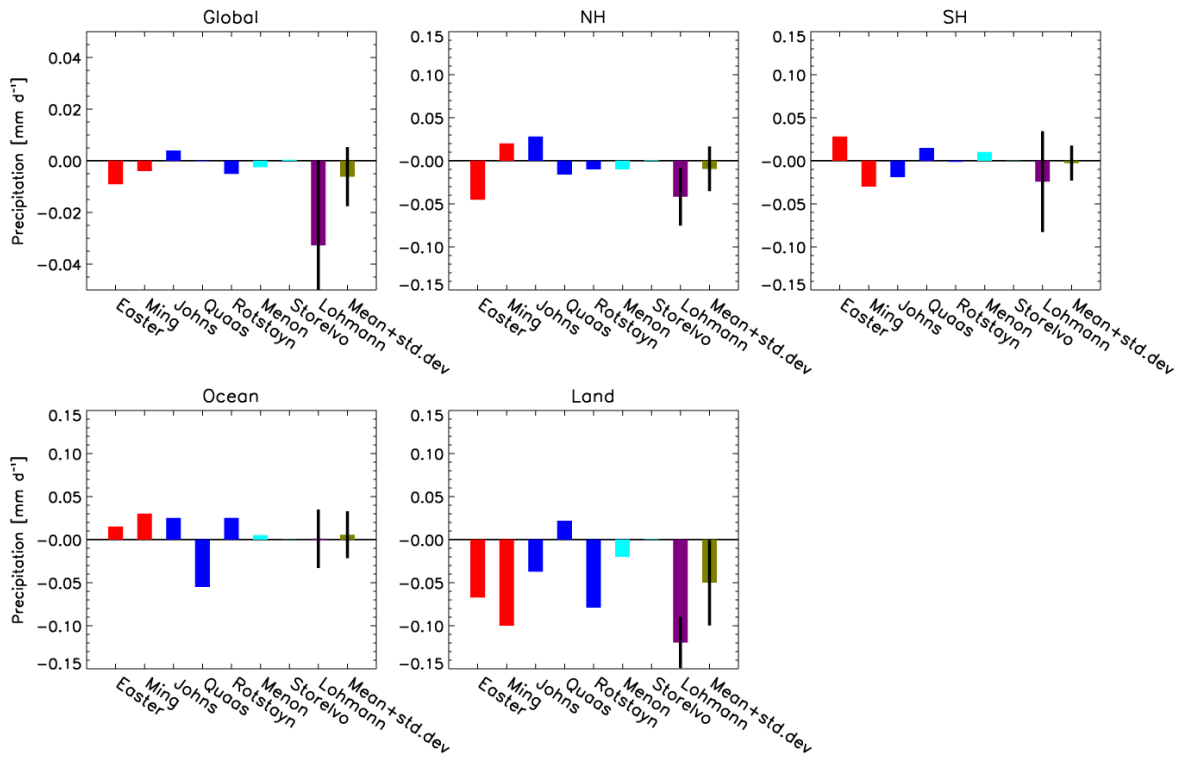


3  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13  
14  
15  
16  
17  
18  
19

**Figure 7.5.3.** Global mean total anthropogenic aerosol effect (direct, semi-direct and indirect cloud albedo and lifetime effects) defined as the change in net radiation at the top-of-the-atmosphere from pre-industrial times to present-day and its contribution over the Northern Hemisphere (NH), Southern Hemisphere (SH), over oceans and over land, and the ratio over oceans/land. Red bars refer to anthropogenic sulphate (Easter et al., 2004; Ming et al., 2005<sup>+</sup>), green bars refer to anthropogenic sulphate and black carbon (Kristjánsson, 2002<sup>\*,+</sup>), blue bars to anthropogenic sulphate and organic carbon (Johns et al., 2004<sup>+</sup>; Quaa et al., 2004<sup>\*,+</sup>; Rotstajn and Liu, 2005<sup>+</sup>), turquoise bars to anthropogenic sulphate, black, and organic carbon (Menon and Del Genio, 2005; Storelvmo et al., 2005<sup>\*</sup>), dark purple bars to the mean and standard deviations of anthropogenic sulphate, black, and organic carbon effects on water and ice clouds (Lohmann and Diehl, 2005), teal bars refer to a combination of ECHAM4 GCM and POLDER satellite results (Lohmann and Lesins, 2002) and olive bars to the mean plus standard deviation from all simulations.

\* refers to estimates of the aerosol effect deduced from the shortwave radiative flux only  
+ refers to estimates solely from the indirect effects

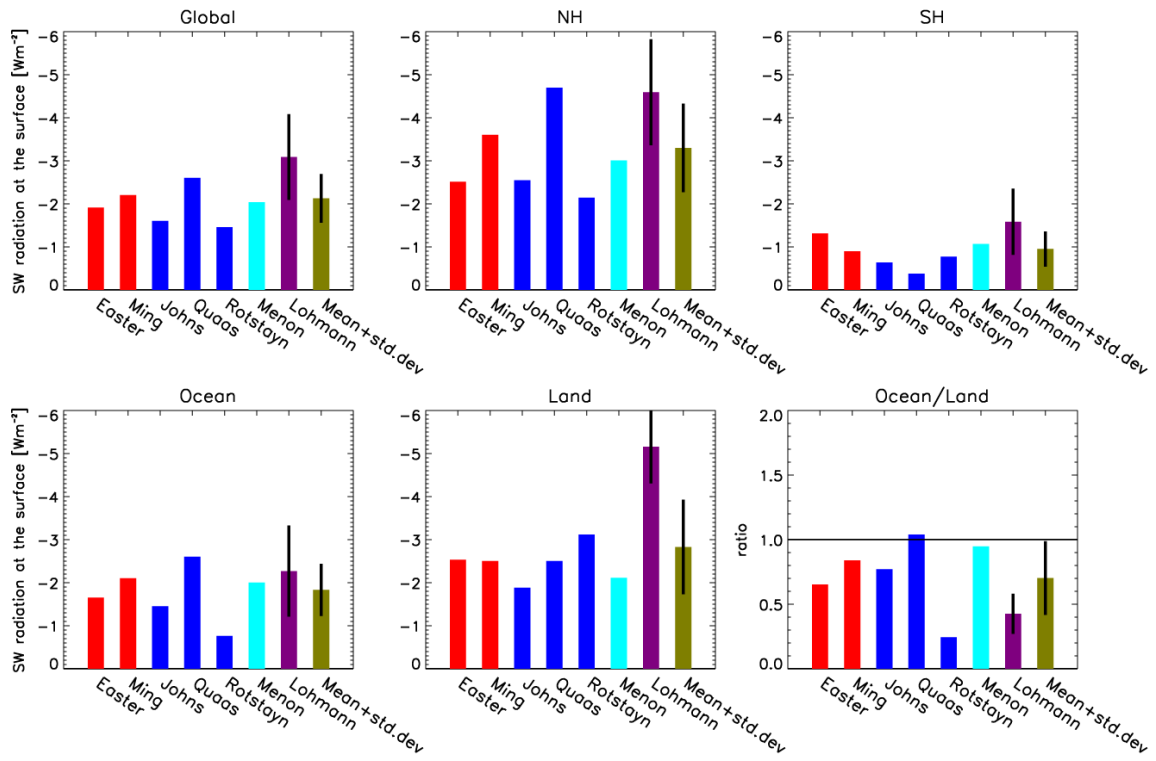
1  
2



3  
4  
5  
6  
7  
8

**Figure 7.5.4.** Same as Figure 7.5.3, but for precipitation. Note the different vertical scale for the global mean precipitation

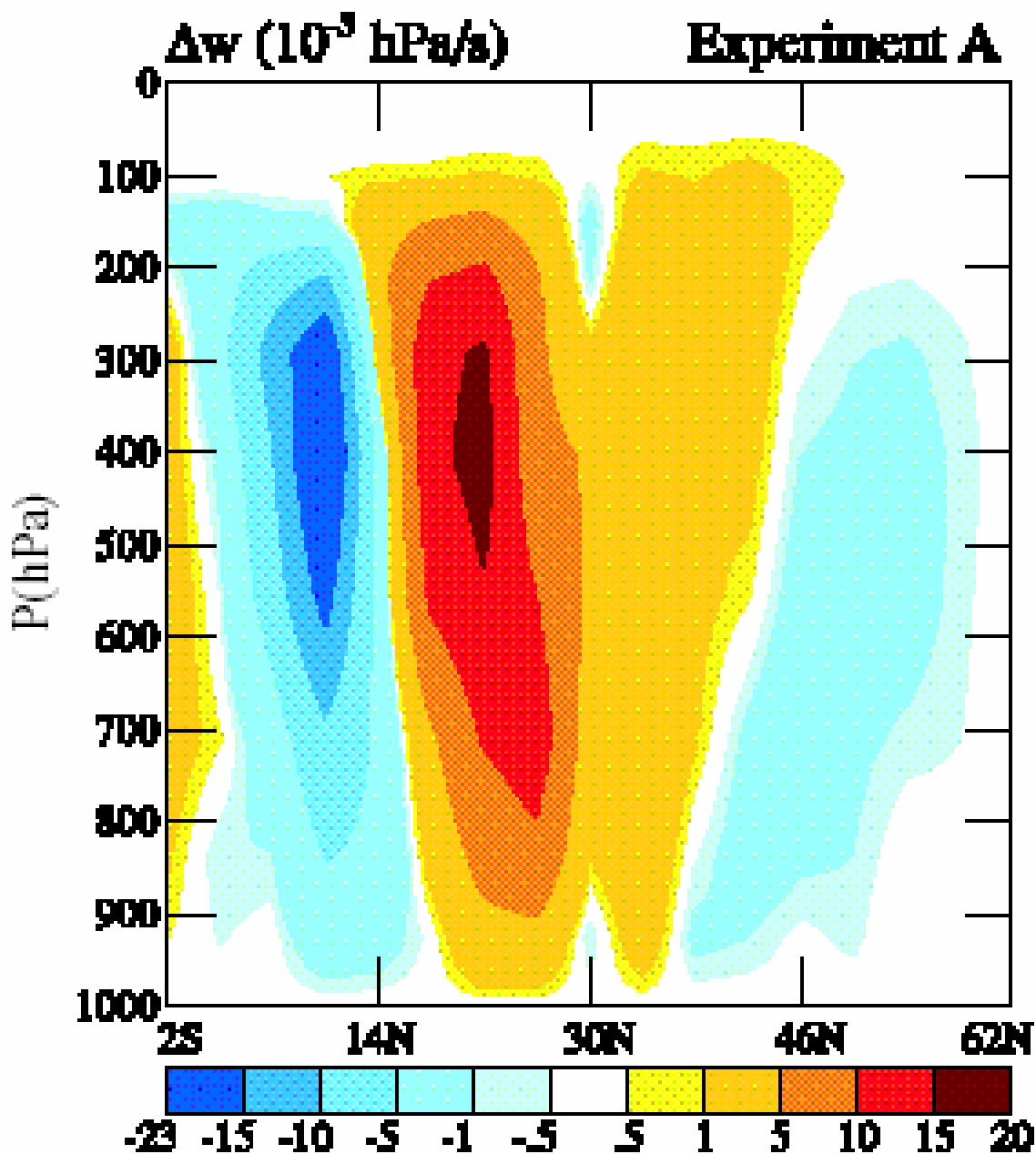
1  
2



3  
4  
5  
6

**Figure 7.5.5.** Same as Figure 7.5.3, but for the net shortwave radiation at the surface.

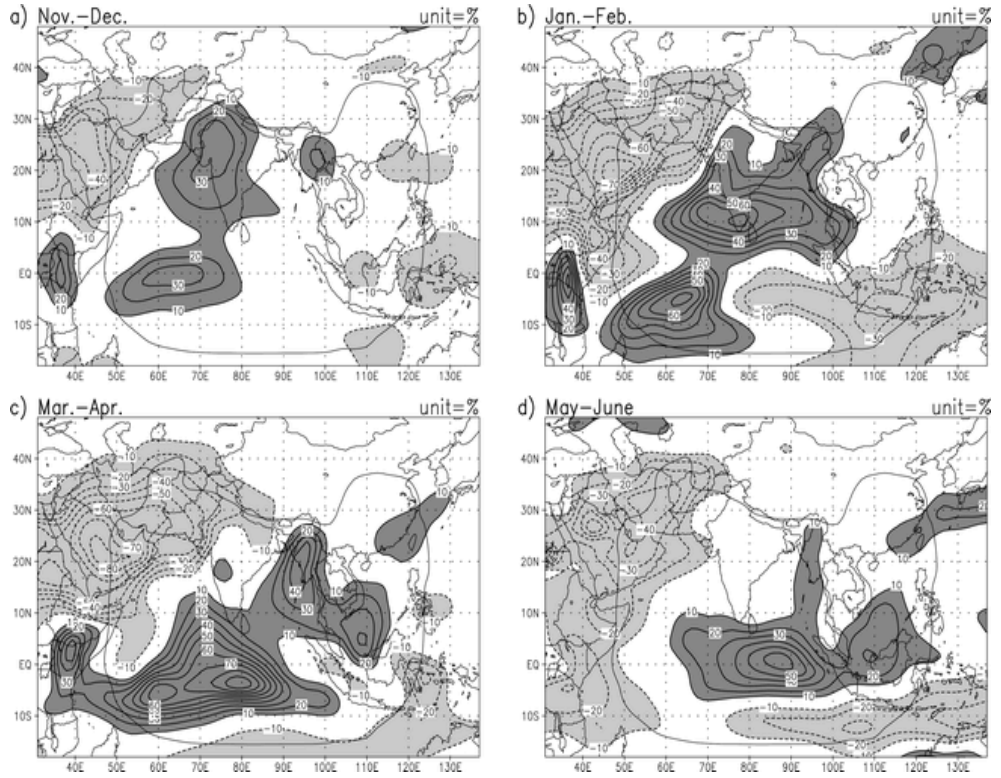
1  
2



3  
4  
5  
6  
7  
8  
9

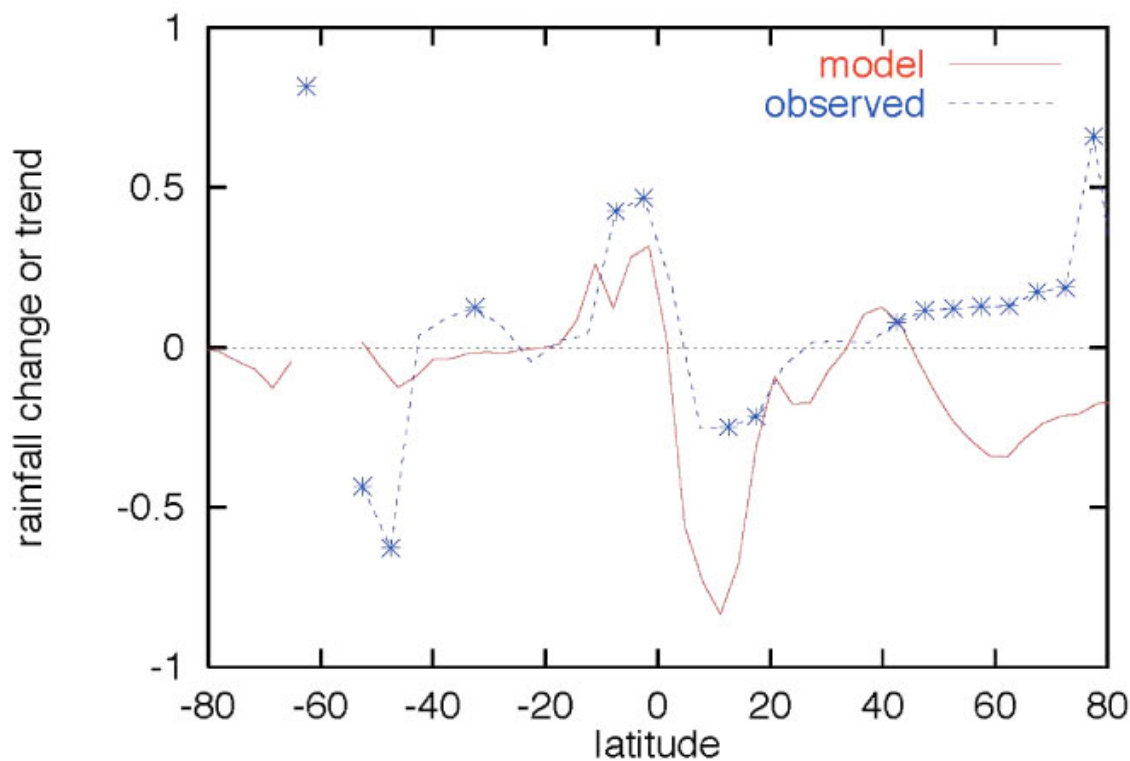
**Figure 7.5.6.** Simulated (GISS GCM) JJA vertical velocity change as a function latitude and altitude averaged over 90°E to 130°E for the experiment with aerosols representative of the measurements made over the Indian Ocean region and industrial regions of China (With courtesy from Menon et al., 2002b).

1  
2

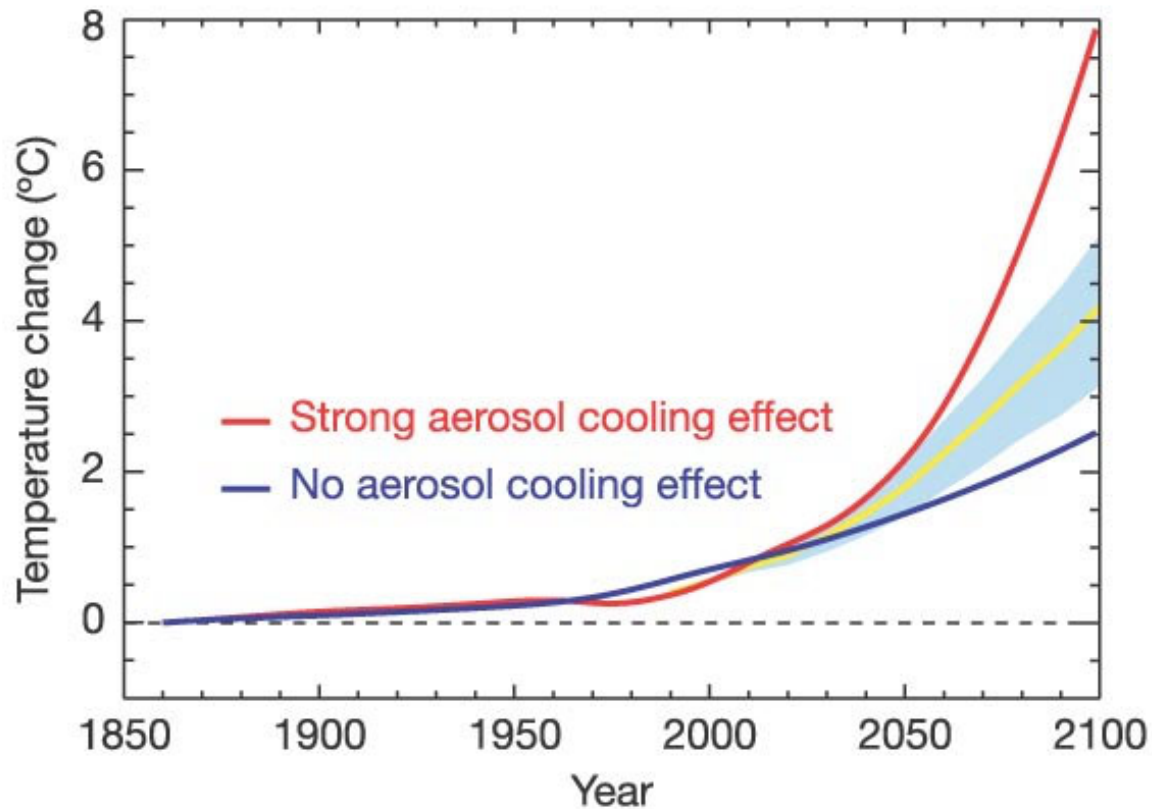


3  
4  
5  
6  
7  
8  
9  
10

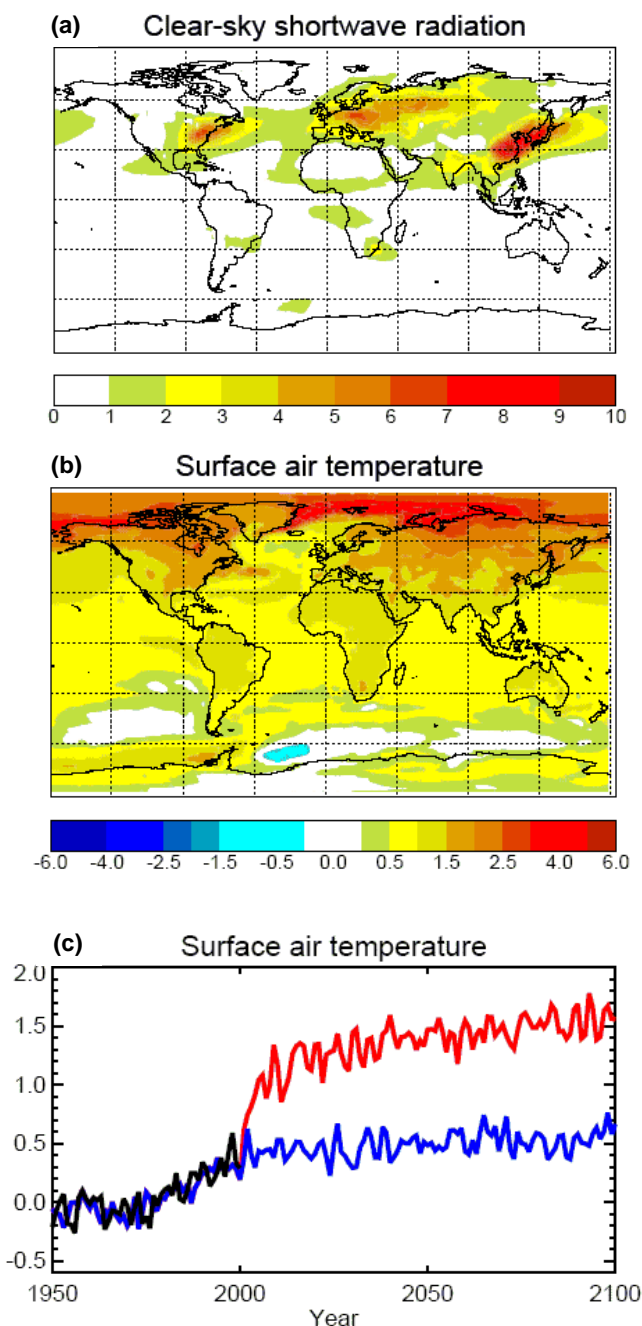
**Figure 7.5.7.** Percentage change in precipitation due to imposed aerosol forcing over the Indian Ocean region for a ratio of surface to atmospheric forcing of  $-0.9$ . The ellipse-like contour in the figure denotes the area where the forcing in the atmosphere exceeded  $3 \text{ W m}^{-2}$  (With courtesy from Chung et al., 2002).

1  
23  
4  
5  
6  
7  
8  
9  
10  
11

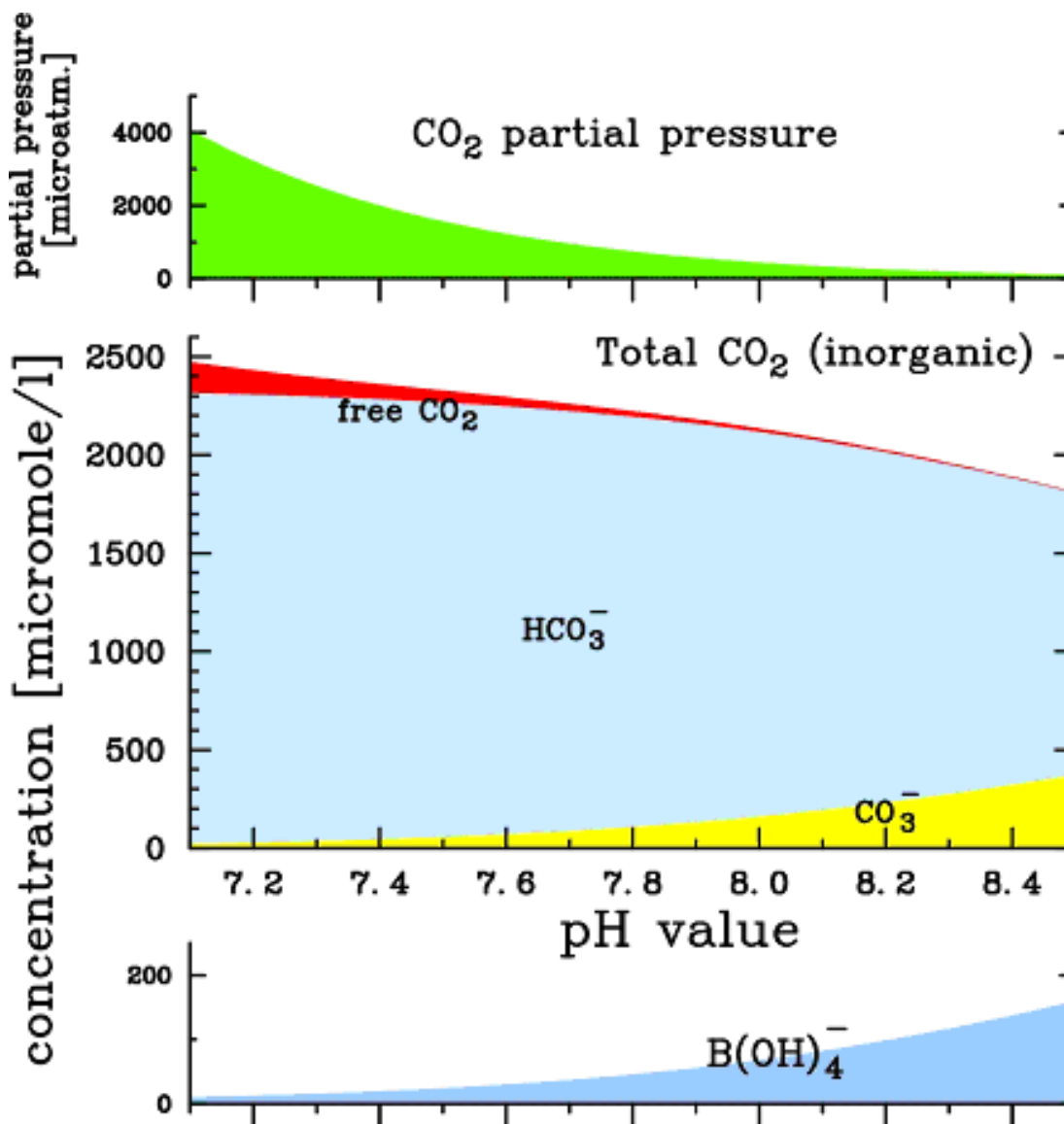
**Figure 7.5.8.** Zonally averaged trend in observed annual-mean precipitation over land for the period 1901–1998 after Hulme et al. (1998) in  $\text{mm day}^{-1}$  per century (blue dotted line) and zonally averaged difference in annual-mean precipitation over land between the present-day and pre-industrial simulations in  $\text{mm day}^{-1}$  (solid red line). Points at which the observed trend is (is not) significant at the 5% level are shown as asterisks (pluses). Courtesy of Rotstayn and Lohmann (2002).

1  
23  
4  
5  
6  
7  
8  
9  
10  
11

**Figure 7.6.1.** Temperature change simulated by Andreae et al. (2005) for the period 1850 to 2100. Two extreme cases are shown: strong present-day aerosol cooling consistent with 'forward' studies of aerosol effects on climate but with a climate sensitivity not ruled out by observations (red line,  $Q_{\text{aeros}} = -1.7 \text{ W m}^{-2}$ ), and the case of no aerosol cooling effect (blue line). The shading and the yellow line represent the range and central projection given in IPCC-TAR, based on the same scenario used in these calculations (scenario A2).

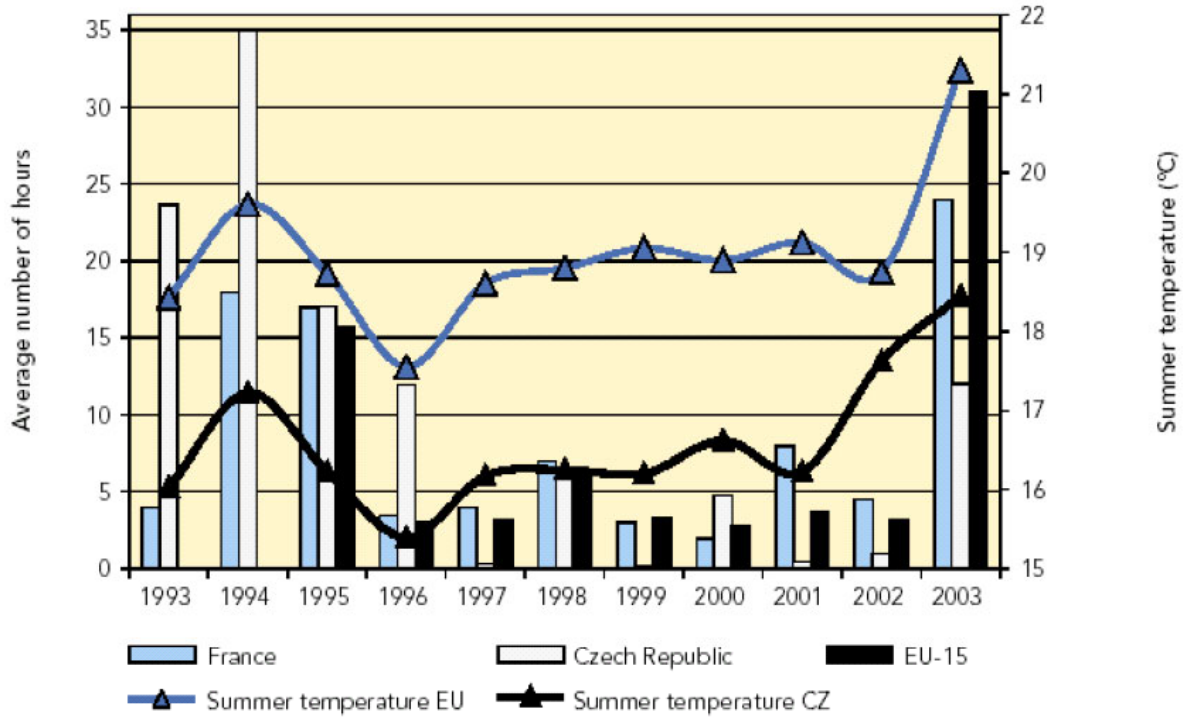
1  
23  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13  
14  
15

**Figure 7.6.2.** Effect of removing the entire burden of sulphate aerosols in year 2000 on (upper panel) the annual mean clear-sky top of the atmosphere shortwave radiation ( $\text{W m}^{-2}$ ) calculated by Brasseur and Roeckner (2005) for the time period 2071–2100, and (middle panel) on the annual mean surface air temperature ( $^{\circ}\text{C}$ ) calculated for the same time period. Lower panel: temporal evolution of global and annual mean surface air temperature anomalies ( $^{\circ}\text{C}$ ) with respect to the mean 1961–1990 values. The evolution prior to year 2000 is driven by observed atmospheric concentrations of greenhouse gases and aerosols as adopted by IPCC (see Chapter 10). For years after 2000, the concentration of greenhouse gases remains constant while the aerosol burden is unchanged (blue line) or set to zero (red line). The black curve shows observations (Jones et al., 2001: Global and hemispheric temperature anomalies 1856–2000 – land and marine instrumental records. <http://cdiac.ornl.gov/trends/temp/jonescru/jones.html>).

1  
23  
4  
5  
6  
7  
8  
9

**Box 7.1, Figure 1.** The equilibrium partitioning of the different carbon species in seawater (at alkalinity 2350  $\mu\text{mol L}^{-1}$ , temperature 15°C, salinity 35): A higher CO<sub>2</sub> partial pressure is directly coupled with a lower pH value, i.e., more acid conditions.

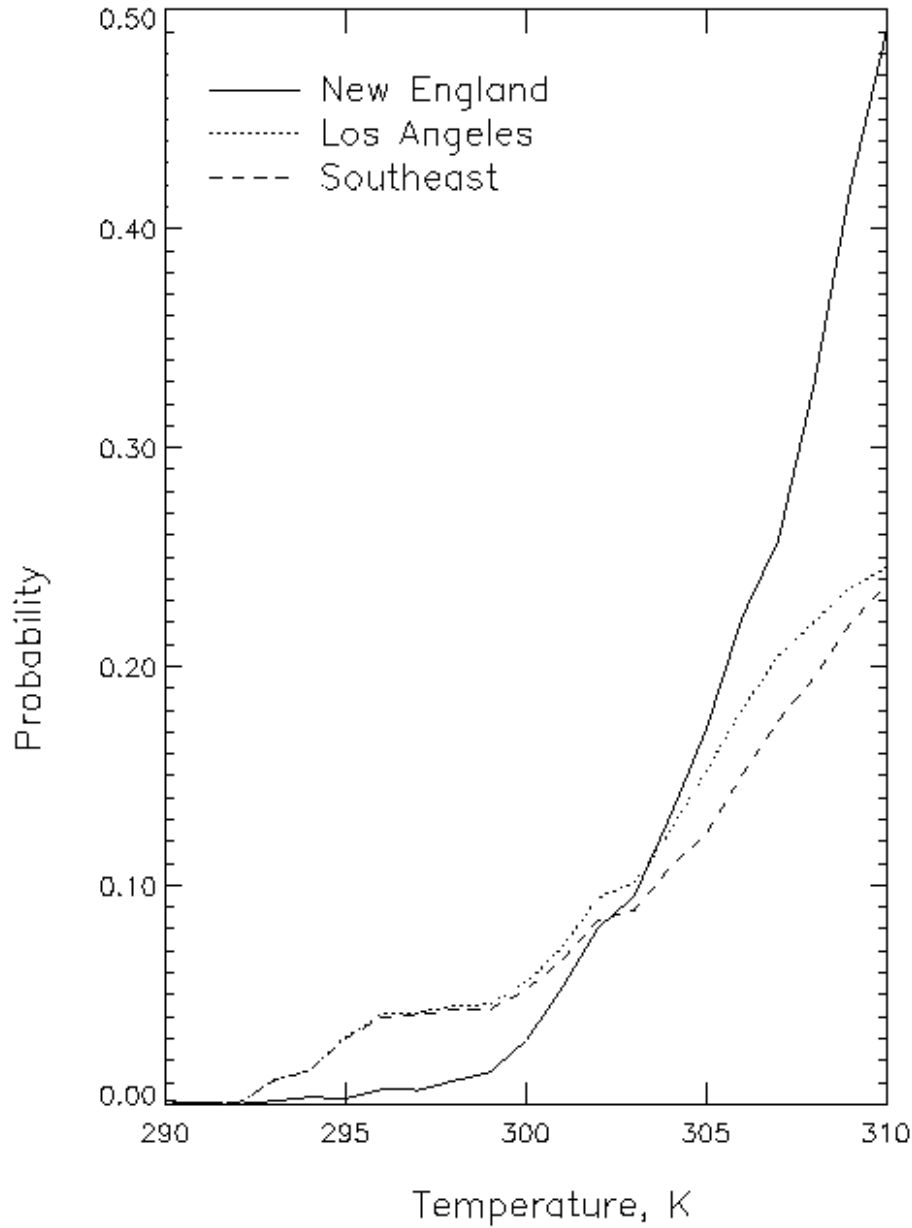
1  
2



3  
4  
5  
6  
7  
8  
9

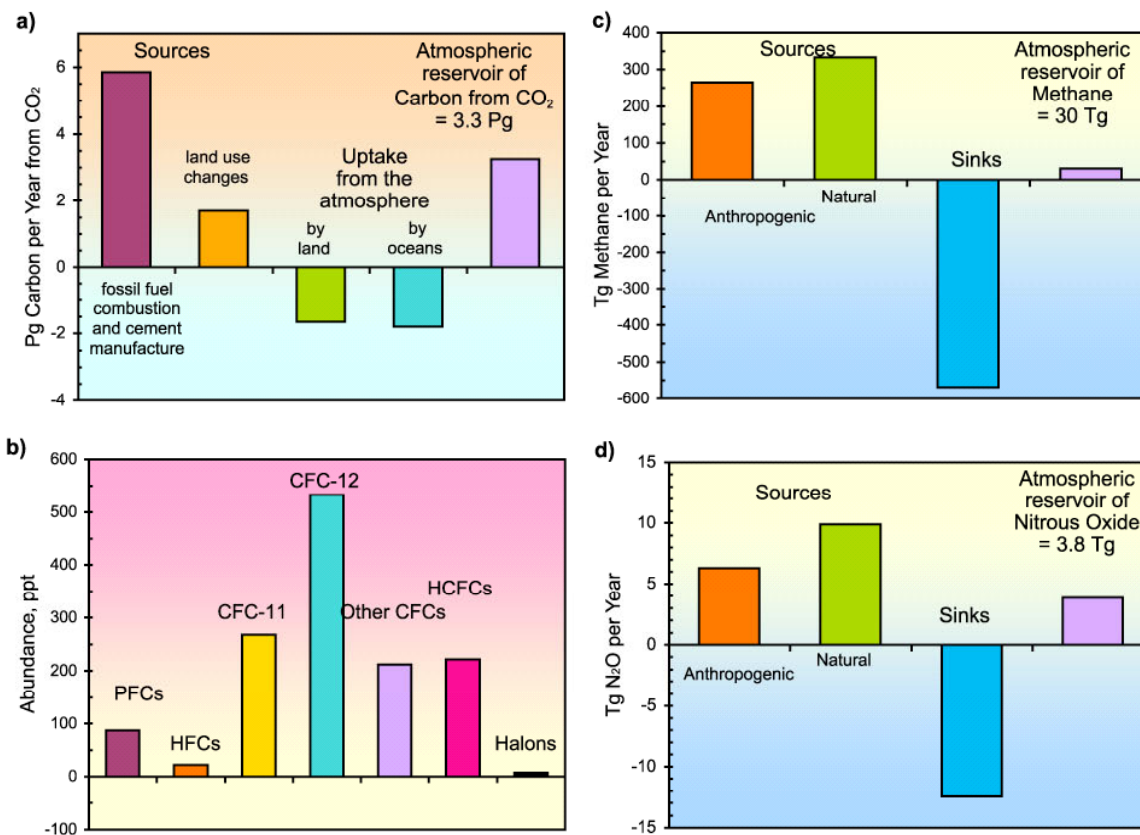
**Box 7.2, Figure 1.** Average number of hours with ozone concentrations exceeding 180 µg/m³ in France, the Czech Republic (CZ), and the European Union for the period 1993–2003, illustrating the strong link with temperature. From Science Panel on Atmospheric Research (2005).

1  
2



3  
4  
5  
6  
7  
8  
9  
10  
11

**Box 7.2, Figure 2.** Probability that the daily maximum 8-hour average ozone will exceed the U.S. National Ambient Air Quality Standard (NAQS) of 0.08 ppmv for a given daily maximum temperature, based on 1980-1998 data. Values are shown for New England (bounded by 36N, 44N, 67.5W, and 87.5W), the Los Angeles Basin (bounded by 32N, 40N, 112.5W, and 122.5W) and the southeastern United States (bounded by 32N, 36N, 72.5W, and 92.5W). From Lin et al. (2001).

1  
23  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13  
14  
15  
16  
17

**Question 7.1, Figure 1.** Breakdown of contributions to the changes in atmospheric greenhouse-gas concentrations. (a) Human contributions to atmospheric carbon dioxide (CO<sub>2</sub>) for 1980–2000. Each year carbon dioxide is released to the atmosphere by human activities including fossil fuel combustion and land use change. Not all of the carbon dioxide emitted remains in the atmosphere. Some is dissolved into the oceans and some is incorporated into plants as they grow. 1 Pg = 10<sup>15</sup>g. (b) Atmospheric abundances in 1998 of CFCs and other halogen-containing compounds. These chemicals are exclusively human-produced. 1 ppt = 1 part in 10<sup>12</sup>. (c) Sources and sinks of methane (CH<sub>4</sub>). Anthropogenic or human-caused sources of methane include emission of methane from energy production, land fills, raising ruminant animals, rice agriculture and biomass burning. Tg = 10<sup>12</sup>g. (d) As (c), but for nitrous oxide (N<sub>2</sub>O). Anthropogenic or human-caused sources of N<sub>2</sub>O include the transformation of fertilizer N into N<sub>2</sub>O and its subsequent emission from agricultural soils, biomass burning, emissions from cattle and feedlots, and some industrial sources, including nylon manufacture.