

Chapter 7: Clouds and Aerosols

Coordinating Lead Authors: Olivier Boucher (France), David Randall (USA)

Lead Authors: Paulo Artaxo (Brazil), Christopher Bretherton (USA), Graham Feingold (USA), Piers Forster (UK), Veli-Matti Kerminen (Finland), Yutaka Kondo (Japan), Hong Liao (China), Ulrike Lohmann (Switzerland), Philip Rasch (USA), S. K. Satheesh (India), Steven Sherwood (Australia), Bjorn Stevens (Germany), Xiao-Ye Zhang (China)

Contributing Authors: Govindswamy Bala (India), Nicolas Bellouin (UK), Cristina Facchini (Italy), Mark Flanner (USA), Steve Ghan (USA), Claire Granier (France), Corinna Hoose (Germany), Makoto Koike (Japan), Natalie Mahowald (USA), Gunnar Myhre (Norway), Alan Robock (USA), Bjørn Samset (Norway), Hauke Schmidt (Germany), Michael Schulz (Norway), Trude Storelvmo (USA)

Review Editors: Sandro Fuzzi (Italy), Joyce Penner (USA), Venkatachalam Ramaswamy (USA), Claudia Stubenrauch (France)

Date of Draft: 16 December 2011

Notes: TSU Compiled Version

Table of Contents

Executive Summary	3
7.1 Introduction	5
7.2 Clouds	6
7.2.1 <i>Clouds in the Present-Day Climate System</i>	7
7.2.2 <i>Process Modelling and Observation of Clouds</i>	9
7.2.3 <i>Representation of Clouds in Climate Models</i>	10
7.2.4 <i>Cloud and Water-Vapour Feedback</i>	13
7.2.5 <i>Basis of Precipitation Changes in Cloud Physical Processes</i>	19
7.2.6 <i>Anthropogenic Sources of Cloudiness</i>	21
7.3 Aerosols	22
7.3.1 <i>Introduction</i>	22
7.3.2 <i>Aerosol Sources and Processes</i>	22
7.3.3 <i>Progresses and Gaps in Understanding Climate Relevant Aerosol Properties</i>	24
7.3.4 <i>Aerosol Distributions</i>	28
7.3.5 <i>Aerosol Radiative Effects</i>	30
7.3.6 <i>Aerosol-Climate Feedbacks</i>	35
7.4 Aerosol-Cloud Interactions	37
7.4.1 <i>Introduction</i>	37
7.4.2 <i>Aerosol Effects on Liquid Cloud Albedo (Indirect Radiative Forcing – iRF)</i>	39
7.4.3 <i>Adjustments in Liquid Clouds</i>	41
7.4.4 <i>Adjustments in Cold Clouds</i>	43
7.4.5 <i>Aerosol-Cloud Microphysical Effects on Precipitating Systems</i>	45
7.4.6 <i>Synthesis of Aerosol Effects</i>	48
7.4.7 <i>Impact of Cosmic Rays on Aerosols and Clouds</i>	50
7.5 Solar Radiation Management and Related Techniques	52
7.5.1 <i>Introduction</i>	52
7.5.2 <i>Idealised Experiments</i>	53
7.5.3 <i>Stratospheric Aerosols</i>	53
7.5.4 <i>Cloud Brightening</i>	54
7.5.5 <i>Surface Albedo Changes</i>	55
7.5.6 <i>Cirrus Thinning</i>	55
FAQ 7.1: How do Aerosols Affect Climate and Climate Change?	56

1 **FAQ 7.2: How do Clouds Affect Climate and Climate Change?57**
2 **FAQ 7.3: Could Geoengineering Counteract Climate Change and What Side-Effects Might Occur?..58**
3 **References.....61**
4 **Tables100**
5 **Figures102**
6

Executive Summary

- Clouds cool the Earth on average, by about 17 W m^{-2} . This is the net result of a greenhouse (infrared) warming due mainly to high clouds ($\sim 30 \text{ W m}^{-2}$) and a cooling effect from reflecting solar radiation contributed by all cloud types ($\sim 47 \text{ W m}^{-2}$).
- New satellite observations and advances to models have given us global simulations that can explicitly resolve some types of clouds. Comparison to detailed observations with such models has led to improved understanding of cloud interactions with the meteorology and the climate. However observations alone do not provide a robust constraint on the sign and magnitude of cloud feedbacks.
- Evidence for a net positive feedback from water vapour and lapse rate changes has increased robustness. The net long-term feedback parameter is very likely positive with an interquartile range of 0.90 to $1.06 \text{ W m}^{-2} \text{ K}^{-1}$ from CMIP3 model [PLACEHOLDER FOR SECOND ORDER DRAFT: CMIP5 models].
- Cloud feedbacks on long-term greenhouse-gas induced surface temperature change are likely positive. Robust positive feedback mechanisms have been established while no mechanism for strong negative global cloud feedback has convincing observational or model-based support. The robust mechanisms include a rise in the heights of cirrus cloud tops and a reduction in subtropical cloudiness. The range of the cloud feedback parameter in the CMIP5 models is [xx to yy] $\text{W m}^{-2} \text{ K}^{-1}$. Inconsistent prediction of changes to low cloud remains the largest source of spread in the cloud feedback parameter and equilibrium climate sensitivity among climate models. Since all cloud types are crudely represented by climate models, values outside the current spread of climate models cannot be ruled out.
- Observations, theoretical considerations and models indicate that the strength of extreme precipitation events, which can cause flooding, tend to strongly increase as the climate warms and atmospheric humidity increases. This tendency is expected to vary significantly by region.
- Observational studies since AR4 suggest that contrails and contrail-cirrus from aircraft, at current levels of coverage, exert only a small adjusted forcing (AF) of 0.03 W m^{-2} (with a 5%–95% uncertainty range of 0.01 to 0.06 W m^{-2}), and are unlikely to have an observable effect on surface temperature and diurnal temperature range.
- There has been continuous progress since AR4 on observing and modelling climate-relevant aerosols properties (including their size distribution, hygroscopicity, chemical composition, mixing state, optical and cloud nucleation properties) and their atmospheric distribution. The representation of aerosol processes in the CMIP5 models remain more simplistic than in some of the more detailed aerosol models used to assess radiative forcing (RF).
- The aerosol direct effect was assessed for the year 2010 relative to 1750 with all ranges indicative of 5%–95% confidence intervals. The RF for the total direct aerosol effect is $-0.3 \pm 0.3 \text{ W m}^{-2}$ (not accounting for possible anthropogenic changes in mineral dust), using evidence from aerosol models and some constraints from observations. The semi-direct effect of atmospheric heating on clouds can be significant and of either sign regionally but is thought to be small globally ($\sim 0.1 \text{ W m}^{-2}$) and not significantly different than zero. The direct and semi-direct effects from anthropogenic aerosols are assessed together as an AF of $-0.3 \pm 0.4 \text{ W m}^{-2}$.
- Sulphate aerosol is responsible for a RF of -0.3 W m^{-2} (-0.2 to -0.6 W m^{-2}). Black carbon (BC) aerosol has a RF $+0.2 \pm 0.2 \text{ W m}^{-2}$ (fossil fuel sources only) and $+0.4 \pm 0.2 \text{ W m}^{-2}$ (fossil fuel and biomass burning including a possible small fraction from vegetation feedbacks). The largest uncertainties relate to the vertical profile of BC. Organic carbon aerosol from fossil fuel sources has a RF $-0.05 \pm 0.05 \text{ W m}^{-2}$. Biomass burning aerosol has a RF -0.01 W m^{-2} (-0.15 to $+0.1 \text{ W m}^{-2}$). Secondary organic aerosol has a RF -0.04 W m^{-2} (-0.03 to -0.07 W m^{-2}) and nitrate aerosol has a RF of $-0.1 \pm 0.08 \text{ W m}^{-2}$. Finally mineral aerosol has a RF of $-0.1 \pm 0.2 \text{ W m}^{-2}$ but this may include part of a feedback.
- Anthropogenic absorbing aerosols (BC and brown carbon) on snow and ice are responsible for a positive RF of $+0.04 \text{ W m}^{-2}$, with a 0.01– 0.10 W m^{-2} (5%–95%) uncertainty range. This radiative forcing is 2–4

1 time more effective at causing global mean temperature changes than an equivalent radiative forcing from
2 CO₂.

- 3
- 4 • A number of climate feedbacks have been identified which involve aerosols, either through a change in
5 the source strength of natural aerosols or a change in sink processes. There is low agreement in model
6 simulations and no solid evidence to suggest that such feedbacks could be significant during the 21st
7 century although they may be important at the regional scale.
- 8
- 9 • There has been continuous progress in our understanding of aerosol-cloud interactions in liquid clouds, in
10 particular from observations and large-eddy simulating models, which reveal some compensating effects.
11 Progress has been made on our understanding of aerosol interactions with mixed phase and ice clouds and
12 their representation in climate models. This progress has led to a reduction in the estimate for the
13 magnitude of global aerosol indirect forcings.
- 14
- 15 • The indirect radiative forcing (iRF) is very likely between -1 and -0.1 W m^{-2} , the lower bound being
16 based on estimates from climate models, and likely between -0.4 and -0.2 W m^{-2} , the lower bound being
17 based on studies that take satellite data into account. Following the same line of argumentation, the
18 indirect adjusted forcing (iAF) is very likely between -1.5 and 0 W m^{-2} and likely between -0.7 and -0.2
19 W m^{-2} .
- 20
- 21 • There is limited, if any, evidence and no agreement that the small-scale impact of aerosols on cloud
22 microphysical structure translates into a significant regional impact in terms of precipitation amount
23 (beyond orographic locations) but there is medium evidence and agreement for an effect on timing and
24 intensity of precipitation.
- 25
- 26 • New studies provide robust evidence that cosmic rays influence new particle formation through changes
27 in atmospheric ionization rate. However there is medium evidence and high agreement that any effects
28 from variations in cosmic rays on CCN and cloud properties are insignificant climatically.
- 29
- 30 • From a physical-science assessment basis, model studies, observations of the effects of volcanic
31 eruptions, and physical arguments suggest that some Solar Radiation Management (SRM) strategies for
32 geoengineering may be effective in offsetting the global average surface temperature increase. However
33 SRM would produce an inexact compensation for the RF by greenhouse gases and there would be
34 residual regional differences in temperature and rainfall patterns. SRM will not compensate for ocean
35 acidification from increasing CO₂ and may have other impacts on the climate system (e.g., stratospheric
36 ozone depletion from stratospheric aerosol injection). Termination of SRM would produce a reappearance
37 of most of the avoided global warming within about a decade.
- 38
- 39 • Evidence from past volcanic eruptions and modelling studies suggest that increasing the amount of
40 aerosols in the stratosphere can increase the Earth's albedo enough to counteract the global RF of at least
41 up to a doubling of CO₂ (within the limitations stated above). The effectiveness and potential of SRM
42 through cloud brightening is more uncertain than through stratospheric aerosol injection because of our
43 limited understanding of aerosol indirect effects on clouds.
- 44

7.1 Introduction

The atmosphere, although mostly composed of gases, is full of particles. It is usual to partition these particles into cloud particles, atmospheric aerosols, and falling hydrometeors according to their size, water content and sedimentation velocity.

Clouds usually form in rising air, which expands and cools until cloud formation occurs through nucleation or freezing of aerosol particles. Cloud particles are generally larger than aerosols and mostly composed of water; they are suspended in the atmosphere and collectively form a cloud which is usually a visible body. The evolution of a cloud is governed by the balance between a number of dynamical, radiative and microphysical processes. Cloud particles of sufficient size become falling hydrometeors, which are categorised as drizzle, raindrops, snow crystals and graupel. Clouds affect the climate system by regulating the flow of radiation at the top of the atmosphere, by controlling precipitation, and through additional mechanisms too numerous to list here. Precipitation processes merit special attention not only because the hydrological cycle is important in its own right, and intricately linked to the structure of regional circulation systems, but also because the frequency and distribution of precipitation is an important sink of aerosol particles.

Atmospheric aerosols are relatively small solid and liquid particles in suspension in the air that can be of natural or anthropogenic origin. They interact with solar radiation, through absorption and scattering, and to a lesser extent with terrestrial radiation, through absorption, scattering and emission. Aerosols can serve as cloud condensation nuclei and ice nuclei upon which cloud droplets and ice crystals form. They also play a wider role in biogeochemical cycles in the Earth system, for instance by carrying nutrients to ocean ecosystems.

Cloud and aerosol amounts and properties are extremely variable in space and time. The short lifetime of clouds in the atmosphere often creates relatively sharp cloud edges and rapid horizontal variations in cloud properties, which is much less typical of aerosol layers. While the dichotomy between aerosols and clouds is generally appropriate and useful, it should be appreciated that there can be a continuum in particle size and a continuum between clear and cloudy sky which makes the distinction between aerosols and clouds, or clouds and rain more difficult and sometimes less relevant (Charlson et al., 2007; Koren et al., 2007).

Both clouds and aerosols are a major source of uncertainties in the climate system. Clouds respond to climate forcing mechanisms in multiple ways and individual cloud feedbacks can be positive or negative. The representation of cloud processes in climate models has been recognised for decades as a continuing source of much of the uncertainty surrounding climate change (e.g., Arakawa 1975, 2004; Bony et al., 2006; Cess et al., 1989; Charney, 1979; Randall, 1989), but with the exception of the Charney report (Charney, 1979) clouds have not been a focal point of past assessment reports. Key issues include the representation of both deep and shallow cumulus convection, microphysical processes in ice clouds, and partial cloudiness that results from small-scale variations of cloud-producing and cloud-destroying processes. Inter-model differences in cloud feedbacks constitute by far the primary source of spread of both equilibrium and transient climate responses simulated by the CMIP3 climate models (Dufresne and Bony, 2008) despite the fact that, as discussed by Randall et al. (2007) and also later in this chapter, most models agree on a near-neutral or positive cloud feedback.

Anthropogenic aerosols are responsible for a radiative forcing of climate through their direct effect (the interaction of aerosols with radiation) and their indirect effects (the interaction of aerosols with clouds). Quantification of the direct and indirect effects of anthropogenic aerosols has proven difficult, and is fraught with uncertainties (Haywood and Boucher, 2000; Lohmann and Feichter, 2005). While previous attempts to quantify the probability distribution function for the net anthropogenic radiative forcing from bottom up approaches have found that it is very likely or virtually certain to be positive, the possibility of negative values could not completely be ruled out (Forster et al., 2007; Haywood and Schulz, 2007). Our inability to better quantify non-greenhouse gas radiative forcings, and primarily that associated with atmospheric aerosols, is partly responsible for the uncertainty in observationally-constrained climate sensitivity (Andreae et al., 2005). It was also found that the total anthropogenic forcing is inversely correlated to climate sensitivity in climate models used for CMIP3 (Kiehl, 2007). This is important because, for a given climate model, the uncertainty in aerosol radiative forcing has proportionally more impact on the simulation of the

1 20th century's climate than of future climate projections (Dufresne et al., 2005) because scenarios of
2 anthropogenic aerosol emissions tend to stabilise or decrease in the future (Lamarque et al., 2010).

3
4 Research into cloud-climate interactions has also progressed significantly since the Fourth Assessment report
5 (AR4, IPCC 2007). Examples include additional field programmes (e.g., the Tropical Warm Pool
6 International Cloud Experiment, TWPICE, as described by May et al. (2008)), greatly improved satellite data
7 (e.g., Stephens and Kummerow, 2007), improved cloud parameterisations (e.g., Park and Bretherton, 2009),
8 studies with high-resolution global models (e.g., Sato et al., 2009), and very-high-resolution regional models
9 (e.g., Khairoutdinov et al., 2009).

10
11 The Third Assessment Report (TAR, IPCC 2001) dedicated a chapter to atmospheric aerosols (Penner et al.,
12 2001), while their radiative forcing was assessed in a separate chapter (Ramaswamy et al., 2001). The AR4
13 updated our understanding of the aerosol radiative forcing (Forster et al., 2007), but did not assess all aspects
14 of aerosol influences on climate. Aerosol indirect effects on climate were discussed in Denman et al. (2007).
15 Our capability to observe aerosols has increased substantially since the TAR and the AR4, and so has our
16 understanding of aerosol processes and our modelling capability, from the fine to the large scale. Many
17 climate models now include more complex parameterisations of aerosol and cloud microphysics,
18 incorporating understanding from process-based models. Models are being more thoroughly evaluated
19 against in-situ (e.g., Koch et al., 2009b) and remote-sensing observations (e.g., Huneeus et al., 2011b), and
20 that evaluation feeds back into model development. However an accurate treatment of aerosol processes and
21 their impact on climate at the global scale remains a challenge.

22
23 For the first time in the IPCC WGI assessment reports, clouds and aerosols are discussed together in a single
24 chapter. This offers the opportunity to assess in a unified framework not only cloud feedbacks and aerosol
25 forcings, but also the multiple interactions among aerosols, clouds and precipitation and their relevance for
26 climate and climate change.

27
28 Figure 7.1 provides an overview of the chapter's approach to clouds and aerosols in the context of climate
29 change. *Forcings* associated with forcing agents such as greenhouse gases and aerosols act on global mean
30 surface temperature through the global radiation budget. *Rapid forcing adjustments* (or rapid responses) arise
31 when forcing agents, by altering flows of energy internal to the system, affect cloud cover (or other
32 components of the climate system) and thereby alter the global budget indirectly. Because these adjustments
33 do not operate through changes to global mean surface temperature, which are slowed by the massive heat
34 capacity of the oceans, they are generally rapid and most are thought to occur within a week. *Feedbacks*
35 amplify or damp changes to the global mean surface temperature via its impact on atmospheric variables that
36 affect the global budget. Such a framework offers a clear distinction between the traditional concept of
37 radiative forcing (RF, defined as the instantaneous radiative forcing with stratospheric adjustment only) and
38 the new concept of adjusted forcing (AF, which includes other atmospheric and surface rapid adjustments) as
39 introduced in Chapter 1 and detailed in Section 8.1.1. The aerosol semi-direct and indirect effects, which
40 have been recognised as important for some time, will be quantified here through the concept of adjusted
41 forcing.

42 43 [INSERT FIGURE 7.1 HERE]

44 **Figure 7.1:** Overview of feedback and forcing pathways involving clouds and aerosols. Forcings are represented by
45 dark arrows; forcing agents are boxes with grey shadows, rapid forcing adjustments (or rapid response) are red arrows
46 and feedbacks are other-colored arrows. See text for further discussion.

47
48 The Chapter aims to discuss clouds and aerosols in a comprehensive but climate-focused way. Section 7.2
49 describes our understanding of the role of clouds in climate change. Section 7.3 discusses aerosol properties
50 and how these link up with best estimates of the aerosol direct radiative forcing and their uncertainties.
51 Section 7.4 covers aerosol-cloud interactions and derives estimates for the aerosol indirect forcing. Finally
52 Section 7.5 assesses solar radiation management techniques aimed at cooling the planet as a number of these
53 techniques rely on the modification of aerosols and clouds. Our radiative forcing estimates for the aerosol
54 effects feed into Chapter 8.

55 56 7.2 Clouds

7.2.1 Clouds in the Present-Day Climate System

7.2.1.1 Cloud Formation, Cloud Types, and Cloud Climatology

To form a cloud, air must cool or moisten until it is sufficiently supersaturated with respect to water vapor to nucleate some of the available condensation or freezing nuclei. Clouds form in diverse ways, including large-scale or orographically-driven ascent, small-scale turbulent buoyant ascent, radiative or evaporative cooling, or turbulent mixing of a moist layer. These formation mechanisms support many important cloud types, e.g., cirrus, stratus, or cumulus, organized by large-scale circulations into different climatological cloud regimes (such as subtropical marine stratocumulus) or transient cloud systems (such as tropical or midlatitude cyclones). Each cloud regime or system has a characteristic assemblage of cloud types that contribute to its radiative properties and precipitation characteristics. Such assemblages can be objectively isolated from satellite data (Jakob and Schumacher, 2008; Jakob et al., 2005).

Figure 7.2 shows a selection of widely occurring tropical and extratropical cloud regimes, and how they might look on at visible wavelengths on a typical geostationary satellite image.

[INSERT FIGURE 7.2 HERE]

Figure 7.2: Diverse cloud regimes reflect diverse meteorology. (a) A visible-wavelength geostationary satellite image shows (from top to bottom) expanses and long arcs of cloud associated with extratropical cyclones, subtropical coastal stratocumulus near Baja California breaking up into shallow cumulus clouds in the central Pacific, and mesoscale convective systems outlining the Pacific ITCZ. (b) A schematic vertical section through a typical warm front of an extratropical cyclone shows multiple layers of upper-tropospheric ice (cirrus) and mid-tropospheric water (altostratus) cloud upwind of the frontal zone, an extensive region of nimbostratus associated with frontal uplift and turbulence-driven boundary layer cloud in the warm sector. (c) A schematic cross section along the low-level trade wind flow from a subtropical west coast of a continent to the ITCZ shows typical low-latitude cloud types, shallow stratocumulus in the cool waters of the oceanic upwelling zone near the coast, trapped under a strong subsidence inversion, shallow cumulus of warmer waters further offshore and a transition into precipitating cumulonimbus cloud systems with extensive cirrus anvils associated with rising air motions in the ITCZ.

Figure 7.3a shows a corresponding geographical annual-mean plot of the total fraction of atmospheric columns that contain cloud, thresholded to remove very thin cloud of little radiative significance. Figure 7.3b shows a latitude-height section of annual- zonal-mean cloud fractional occurrence from CloudSat and Calipso, a combination of active sensors that can detect even thin clouds (optical depth less than roughly 3–5) and see deep into thick cloud layers. Clouds appearing above roughly the 400 hPa level (which are nearly all ice) are typically considered “high clouds”, while those appearing below roughly 700 hPa (which are mostly liquid but often contain ice outside the tropics) are considered “low” (Zelinka et al., 2011a).

[INSERT FIGURE 7.3 HERE]

Figure 7.3: Annual-mean cloud fractional occurrence based on four years of satellite observations (June 2006–February 2011) from CloudSat and Calipso (Kay et al., 2011; COSP simulator). (a) Geographical mean, with thin cloud ($SR < 5$) removed; (b) latitude-height section of zonal mean cloud cover. [PLACEHOLDER FOR SECOND ORDER DRAFT: further graphical refinement.]

[INSERT FIGURE 7.4 HERE]

Figure 7.4: Distribution of annual-mean SWCRE, LWCRE, net CRE (from CERES-EBAF) and precipitation (from CMAP).

7.2.1.2 Effects of Clouds on Earth’s Radiation Budget

The effect of clouds on Earth’s radiation budget can be inferred by comparing satellite estimates of upwelling solar and longwave radiation with the corresponding fluxes in the absence of clouds, obtained by screening cloudy pixels or using clear-sky radiative transfer models. Using this method, Loeb et al., (2009) estimate that in annual global mean, clouds enhance the planetary albedo, changing the net shortwave radiative flux into the top of the atmosphere (TOA) by -47 W m^{-2} compared to a cloud-free atmosphere (the global shortwave cloud radiative effect or SWCRE), and that clouds enhance the planetary greenhouse effect, reducing net longwave radiative energy loss by 30 W m^{-2} (the global longwave cloud radiative effect or LWCRE). Other published estimates differ from these by 10% or less (Loeb et al., 2009). The combined annual global mean net CRE of -17 W m^{-2} can be regarded as a cooling effect of clouds on climate. Both

1 global mean SWCRE and LWCRE are large compared to the 4 W m^{-2} radiative forcing of doubling CO_2 .
2 Hence, even modest changes in the global distribution of clouds could produce substantial radiative feedback
3 on climate change.

4
5 The regional patterns of annual-mean SWCRE and LWCRE, shown in Figure 7.4a-b, reflect typical cloud
6 regimes averaged across the seasonal cycle. High clouds, which are cold compared to the underlying surface,
7 dominate patterns of LWCRE, while the SWCRE is sensitive to optically thick clouds at all altitudes. Figure
8 7.2 shows that regions of deep, thick cloud with large LWCRE and large negative SWCRE tend to
9 accompany precipitation, emphasizing their intimate connection with the hydrological cycle. The net CRE is
10 negative over most of the globe and most negative in regions of very extensive low-lying reflective cloud
11 such as the midlatitude and eastern subtropical oceans, where SWCRE is strong but LWCRE is weak.

12 7.2.1.3 *Coupling Between Clouds, Precipitation, Large-Scale Dynamics and Stratification*

13
14
15 Precipitation is almost exclusively initiated in updrafts inside clouds. Thus, it is inextricably linked to clouds
16 and their dynamical drivers.

17 7.2.1.3.1 *Deep precipitating cloud systems*

18
19 Over the tropical oceans, where most rain falls from cumulus cloud systems, we observe particularly tight
20 statistical correlations over daily or longer timescales and space scales of a few hundred kilometres or more.
21 More rainfall associates with more deep convective cloud cover (Wyant et al., 2006), higher relative
22 humidity throughout the depth of the free troposphere (Holloway and Neelin, 2009) and more mean mid-
23 tropospheric upward motion (Bony et al., 2004; Lintner et al., 2011). Similar relationships have been
24 documented in the midlatitude storm tracks (Norris and Iacobellis, 2005). These relationships may change
25 somewhat if climate changes; for instance, they could be affected by temperature changes that affect
26 saturation water vapour mixing ratio, as well as the depth of the tropopause and the freezing level. However,
27 we do expect a close correspondence between regional changes in mean vertical motion, precipitation and
28 cloud cover that accompany a climate change, a theme explored further in the discussion of cloud feedbacks
29 in Section 7.2.4.

30 7.2.1.3.2 *Large-scale controls on boundary-layer cloudiness*

31
32 While cumulus clouds develop in conditionally unstable layers of the atmosphere, marine stratocumulus
33 clouds persist where there is a strong capping inversion that traps moisture in the atmospheric boundary
34 layer. Klein and Hartmann (1993) showed that on seasonal and longer timescales, increased subtropical
35 marine stratocumulus cloud cover is strongly correlated with greater lower tropospheric stability (LTS)
36 between the surface and 700 hPa. Variants of LTS such as estimated inversion strength (EIS) (Wood;
37 Bretherton 2006) or others that also account for humidity variations (Williams et al., 2006; Zhang et al.,
38 2010a) better predict low cloud cover over the mid-latitude oceans and may be more applicable to changed
39 climates (Wood and Bretherton, 2006).

40 7.2.1.3.3 *Mixed-phase arctic clouds*

41
42 Arctic clouds have become a focus of recent interest because they may affect the sensitivity of the Arctic to
43 climate change, and because they can involve complex, vertically layered interactions between ice, liquid
44 and aerosol particles. These clouds can persist for days, in spite of the inherent instability of the ice-water
45 mix (Fridlind et al., 2007); theory suggests that mixed phase clouds persist if their updrafts are strong
46 enough and ice nucleus concentrations low enough to maintain liquid water and prevent complete glaciation
47 of the cloud (Korolev and Field, 2008). Morrison et al. (2011) argue this leads to a self-regulating structure
48 in which thin layers of liquid water are maintained by radiatively-driven turbulence but are depleted by
49 formation of ice that falls away from the liquid layer. Slight changes in meteorological forcing can rapidly
50 glacialize the cloud layer and greatly diminish its radiative impact (Stramler et al., 2011). This combination of
51 processes challenges numerical models of all scales, which struggle to simulate the balance between liquid
52 and ice particles in field observations of Arctic boundary-layer clouds (Klein et al., 2009).

53
54 The response of high-latitude boundary-layer cloud cover response to the fractional cover of underlying sea-
55 ice could be an important climate feedback and is discussed (including relevant observations) in Section
56 7.2.4.3.5.

7.2.1.3.4 *Small-scale cloud-precipitation-circulation interaction*

Precipitation can feed back profoundly on the small-scale circulations that accompany most cloud systems, for example by generating downdrafts and gusts that can produce more clouds, and by removing condensed water and aerosol particles. This helps foster characteristic ‘open-cell’ patterns of marine boundary layer cloud in midlatitude cold-air outbreaks (e.g., Muller and Chlond, 1996) and the low-latitude trade-wind belts (e.g., Xue et al., 2008); interactions with aerosols may also be involved, allowing rapid transitions between regimes of much vs. little cloud cover (see Section 7.4.3.2). Similar interactions help organize deep cumulus convection into mesoscale convective systems, squall lines, and tornadic thunderstorms (Houze, 1993), and the tendency to organize at these and larger scales gives convection a leading role in driving weather and climate variability especially at low latitudes. The organization is affected by details at small scales such as the fall speed of ice and snow crystals (Houze 1993). These interactions typically occur below the grid scale of climate models, partly explaining why many climate models reproduce convective organization poorly (Mapes et al., 2009).

7.2.2 *Process Modelling and Observation of Clouds*

7.2.2.1 *Challenges in Modelling and Measurement of Cloud Processes*

Cloud formation processes span scales from the submicron scale of cloud condensation nuclei to cloud system scales of up to thousands of kilometres. This range of scales is impossible to cover with direct numerical simulations on computers, and is unlikely to become so for decades if ever.

High-resolution models of individual cloud systems have nonetheless contributed greatly to our appreciation of interactions of turbulence with various types of cloud, e.g., cumulus, stratocumulus and cirrus. The usual strategy is called large-eddy simulation (LES) when applied to boundary-layer turbulence and cloud-resolving modelling (CRM) when applied to deep cumulus convection. The grid spacing is chosen to be small enough to resolve the dominant turbulent eddies that drive cloud heterogeneity, and the effects of unresolved eddies are parameterized. CRMs of deep convective cloud systems with horizontal resolutions of 2 km or finer can skilfully characterize statistical characteristics of the cloud ensemble, including fractional area coverage of cloud, vertical thermodynamic structure, the distribution of updrafts and downdrafts, and organization into mesoscale convective systems. However, some cloud ensemble properties are still sensitive to CRM microphysical parameterization assumptions, particularly the vertical distribution and optical depth of ice and mixed-phase clouds. Also, this type of modelling approach does not capture interactions with large scales, which requires a larger-scale model in which most of the cloud behaviour is parameterized.

Shallow cumulus cloud fields with clouds of below 2 km thickness are widespread over low latitudes. LES of such cloud fields with horizontal grid spacing of ~100 m and vertical grid spacing of ~40 m produces vertical profiles of cloud fraction, temperature, moisture and turbulent fluxes that agree well with available observations, though the simulated precipitation efficiency still shows some sensitivity to microphysical parameterizations (vanZanten et al., 2011).

LES of stratocumulus-topped boundary layers have shown considerable skill in simulating turbulence statistics and vertical thermodynamic structure (e.g., Ackerman et al., 2009; Stevens et al., 2005), and have been used to study the sensitivity of stratocumulus cloud organization, cloud thickness and albedo to changes in cloud condensation nucleus concentration (e.g., Savic-Jovicic and Stevens, 2008; Xue et al., 2008), sea-surface temperature (SST) and free-tropospheric conditions. However, model intercomparisons show that the simulated entrainment rate and stratocumulus cloud thickness are sensitive to the underlying numerical algorithms, even with vertical grid spacings as small as 5 m, due to under-resolution of the sharp capping inversion (Stevens et al., 2005).

7.2.2.2 *Current Observing Capacities Relevant to Cloud Processes and Global Effects*

Observations useful for detecting long-term (at least 20 year) changes in clouds, along with the significant difficulties in interpreting such observations given natural variability and other factors, are discussed in Chapter 2, Section 2.3.8. Here we discuss current observing capabilities for assessing climate-relevant cloud properties. A variety of observing systems, some new since AR4, combine to give us unprecedented insight

1 into different aspects of clouds on different temporal and spatial scales and to test and improve climate
2 model simulations.

3
4 Satellites observations have proved particularly useful in the above regard due to their frequent and global
5 (or near-global) coverage and because they directly measure how cloud characteristics affect outgoing
6 radiation and hence the Earth's energy budget. Instruments available since the late 1970's on a combination
7 of polar orbiters and geostationary weather satellites measure in the visible, near-infrared, thermal and
8 microwave bands of the spectrum, and infer the reflectance, greenhouse effect, and approximate
9 characteristic particle size and liquid water content of clouds. They observe nearly every spot on Earth at
10 intervals from under an hour to many days depending on the instrument. Such satellite observations have
11 limitations - meteorologically different distributions of cloud can look identical to the satellite, particularly if
12 there are multiple cloud layers or cloud properties vary substantially across a satellite pixel (Wolters et al.,
13 2010). Low-lying clouds over ice-covered surfaces are hard to detect in visible channels that are only useful
14 during daytime. Most polar-orbiting satellites overfly each location at two particular local times of day, an
15 issue in many regions with significant diurnal cycles of cloud cover. Geostationary satellites observe
16 throughout the day, but with less spatial resolution.

17
18 Since the late 1990's, other satellites (e.g., NASA's Terra and Aqua) have carried advanced instruments that
19 sample more wavelengths, polarisation, and multiple viewing angles to more accurately estimate the above
20 cloud properties, as well as cloud-top altitudes, and to improve the estimation of rain rates although this
21 remains a challenge (Marchand et al., 2010; Stephens; Kummerow 2007). Since 1997 the TRMM
22 spaceborne radar has also observed the vertical structure of precipitation at latitudes equatorwards of about
23 35°N/S.

24
25 The 2006 launch by NASA of two coordinated, downward-pointing active sensors, the cloud profiling radar
26 (CPR) on the CloudSat satellite and the CALIOP lidar on board the CALIPSO satellite has given the first
27 accurate, near-global picture of the vertical distribution of cloud water and precipitation. Their high
28 sensitivity, small footprint (roughly 1.5 km for CPR and 0.33 km for CALIOP), and vertical resolution (500
29 m and 30 m below 8 km, respectively) enable them to detect more tenuous and smaller clouds than most
30 previous instruments (Winker et al., 2009). Their small daily sampling area makes them most useful for
31 long-term statistics and for improving the interpretation of other instruments with overlapping, but broader,
32 spatial coverage. Since AR4, satellite simulators (Chapter 9) have become widely used for comparing clouds
33 and precipitation simulated by climate models with observations, and new simulators have been developed to
34 compare with modern generations of cloud-observing satellite instruments.

35
36 Surface or aircraft-based observations give detailed process information on clouds in limited regions, often
37 combining many types of measurements. Over the last 15 years, sites in different climate regimes, such as
38 northern Europe, the central USA, China, the tropical Pacific, the West Indies, and the Arctic have begun
39 continuously measuring overlying cloud properties using lidars, radars, and radiometers within a rich context
40 of other atmospheric measurements. Such observations are widely compared with climate model behaviour
41 (e.g., Phillips et al., 2004). Many field programs have been designed to improve understanding of cloud
42 processes important to climate models using multi-instrument, multiplatform observations over a period of a
43 few weeks that sample relationships between many quantities, followed by organized comparisons of climate
44 and process models with the gathered data to evaluate and improve specific aspects of the simulations. Two
45 recent examples are TWP-ICE for tropical cumulus convection in the Australian monsoon and cirrus
46 formation (May et al., 2008) and VOCALS for aerosol-cloud-precipitation interaction in subtropical
47 southeast Pacific stratocumulus clouds (Wood et al., 2011).

48 **7.2.3 Representation of Clouds in Climate Models**

49 **7.2.3.1 Challenges of Parameterization Interaction and Subgrid Variability**

50
51 Clouds form where rising air supersaturates sufficiently with respect to water vapour. The cloud droplets or
52 ice crystals can then evolve, collide and grow to form precipitation. If these processes occurred uniformly
53 across model grid cells, representing clouds in climate models would reduce to parameterizing their
54 microphysics. Especially for ice and mixed-phase clouds this is already a challenge due to the complexity of
55 microphysical processes.
56
57

1
2 Unfortunately clouds are often thin and short-lived, typically form through turbulent processes not resolved
3 by the grid of a general circulation model (GCM), and may vary considerably within a GCM grid cell
4 especially in convective systems or over mountainous terrain. Most CMIP5 climate model simulations use
5 horizontal resolutions of 100–200 km in the atmosphere, with vertical layers varying between 100 m near the
6 surface to more than 1000 m aloft. Within regions of this size in the real world, there is often enormous
7 subgrid variability in cloud properties, associated with variability in humidity, temperature and vertical
8 motion.

9
10 Because of this, the simulation of clouds in most modern climate models involves many interacting
11 parameterizations that must work together as a system. These include parameterization of turbulence and
12 cumulus convection, cloud fraction at each level, vertical overlap of these quantities at different heights, as
13 well as cloud microphysics and aerosol and chemical transport. Each parameterization makes simplifying
14 mathematical assumptions about the nature of subgrid variability within each grid cell; for pragmatic and
15 historical reasons, these assumptions are frequently not fully consistent across the parameterizations used in
16 one model, and vary significantly from model to model. For example, clouds in a grid column may be
17 assumed to be vertically stacked for the radiation calculation, but not for calculating evaporation of
18 precipitation. In summary, realistic simulation of clouds and their response to climate change forms one of
19 the greatest challenges of climate modelling.

20 21 *7.2.3.2 Advances in Microphysical Representation of Liquid Clouds Since AR4*

22
23 Most microphysics schemes used in CMIP5-class climate models no longer assume that clouds are uniform
24 within a model cell, but account for anticipated heterogeneity through simplifying assumptions and concise
25 representations of subgrid-scale variability and covariance of relevant variables. Climate modellers have
26 generally decomposed liquid clouds through a variety of attributes: 1) by considering gross categories of
27 drop size (small cloud drops versus larger precipitating raindrops); 2) by classifying whether the drops
28 developed within convective cloud cores or stratiform clouds that flow out of convective cores or form
29 independently of them; 3) by predicting one or two parameters of an assumed drop size distributions; and/or
30 4) by assuming particular forms (e.g., Gaussian, top-hat) for the spatial variability of fields within a model
31 cell or a cloud.

32
33 Most AR4 era climate models used a bulk (single moment) formulation for stratiform clouds, predicting only
34 the time evolution of the average cloud and rain water mass in each gridbox. Two of 23 CMIP3 models
35 (Storelvmo et al., 2006) employed two-moment formulations for cloud droplets with explicit prognostic
36 equations for cloud drop number concentration (CDNC); others diagnosed CDNC from prognosed aerosols
37 to allow for aerosol indirect effects (Storelvmo et al., 2009), using empirical relationships connecting aerosol
38 mass to CDNC (e.g., Boucher and Lohmann, 1995; Menon et al., 2002), or based on proximity to land and
39 altitude. The two moment schemes employed evolution equations for CDNC that activate cloud drops based
40 on a subgrid vertical velocity and the size spectra of hygroscopic aerosols (Abdul-Razzak and Ghan, 2000;
41 Ghan et al., 2011b; Nenes and Seinfeld, 2003). Many AR4 era models were forced to employ an arbitrary
42 lower bound on CDNC to reduce the AIE, which is undesirable (Hoose et al., 2009).

43
44 More models participating in CMIP5 will use two moment schemes for liquid stratiform cloud. with the
45 following advances. Some models include a diagnostic treatment of rain and snow number concentration as
46 well as mixing ratio (Morrison and Gettelman, 2008; Salzmann et al., 2010), allowing the treatment of
47 aerosol-scavenging and inclusion of the radiative effect of snow. Some models include an explicit treatment
48 of subgrid cloud water variability for calculation of the microphysical process rates (Morrison and
49 Gettelman, 2008). Some models no longer have to specify a lower bound on CDNC. Cloud drop activation
50 schemes are becoming more sophisticated as aerosol schemes are becoming more complex, including more
51 realistic accounting for aerosol hygroscopicity and particle size.

52 53 *7.2.3.3 Advances in Microphysical Representation in Mixed Phase and Ice Clouds Since AR4*

54 55 *7.2.3.3.1 Mixed-phase clouds*

56 New representations of the Bergeron-Wegener-Findeisen process in mixed-phase clouds (Lohmann and
57 Hoose, 2009; Storelvmo et al., 2008b) compare the rate at which the pre-existing ice crystals deplete the

1 water vapour (Korolev 2007) with the condensation rate for liquid water driven by vertical updraft speed.
2 Climate models are increasingly representing detailed microphysics, including mixed phase processes, inside
3 convective clouds (Fowler and Randall, 2002; Lohmann, 2008; Song and Zhang, 2011). Such processes can
4 influence storm characteristics like strength and electrification; more studies are needed to assess their
5 importance for climate simulations.

6 7 *7.2.3.3.2 Ice clouds*

8 Although supersaturation with respect to ice is commonly observed in cirrus clouds, only one AR4 GCM
9 (ECHAM) allowed ice supersaturation (Lohmann and Kärcher, 2002). Several global models now predict ice
10 supersaturation (Gettelman et al., 2010; Liu et al., 2007; Salzmann et al., 2010; Tompkins et al., 2007).
11 Tompkins et al. (2007) assume that once an ice cloud forms, the deposition process is sufficiently rapid that
12 supersaturation is removed within a GCM time step. All other global models predict ice supersaturation
13 based on parameterizations of homogeneous and/or heterogeneous freezing rates, which are discussed in
14 Section 7.4.4.

15 16 *7.2.3.4 Advances in Parameterization of Moist Turbulence and Cumulus Convection*

17 Since AR4, parameterizations of cumulus convection and moist turbulence in many numerical weather
18 prediction and climate models have continued to advance, leading to substantial improvement in their
19 simulation of tropical rainfall and boundary-layer cloud and new capabilities for simulating cloud-aerosol
20 interaction.

21
22
23 New ‘adaptive’ treatments of lateral entrainment into deep cumulus updrafts sensitive to environmental
24 humidity or updraft buoyancy and velocity have improved simulations of the Madden-Julian Oscillation
25 (MJO), tropical convectively-coupled waves, and mean rainfall patterns in the ECMWF operational weather
26 forecast model (Bechtold et al., 2008) and the MIROC4 GCM (Chikira and Sugiyama, 2010). An adaptive
27 detrainment parameterization implemented in the Met Office weather forecast model permits a
28 computationally efficient bulk plume to mimic detrainment from a more realistic ensemble of clouds,
29 improving forecast skill (Derbyshire et al., 2011). Various incremental cumulus parameterization changes
30 have improved other climate models, e.g., modification of the deep convective trigger to account for
31 entrainment effects on updraft buoyancy (Neale et al., 2008), combined with a new parameterization of
32 cumulus momentum fluxes (Richter and Rasch, 2008), improved both the tropical mean state and ENSO
33 variability simulated by the CCSM4 climate model.

34
35 Since AR4, more climate models have adopted cumulus parameterizations that calculate the typical vertical
36 velocity in cumulus updrafts (e.g., Donner et al., 2011; Park and Bretherton, 2009), allowing more realistic
37 representations of cloud microphysics and cloud droplet activation, a key issue for global simulation of
38 aerosol-cloud interaction.

39
40 Several global models have adopted new approaches that more closely couple the parameterization of
41 shallow cumulus convection and moist boundary layer turbulence. The eddy-diffusion mass flux (EDMF)
42 scheme of Siebesma et al. (2007), adopted by ECMWF, combines an eddy-diffusion approach for small-
43 scale turbulence with a mass-flux representation for strong non-cloudy and cloudy turbulent updrafts, which
44 can incorporate a cumulus parameterization (Neggers, 2009; Neggers et al., 2009). In the CAM5 GCM, the
45 shallow cumulus scheme of Park and Bretherton (2009) coupled to the turbulence parameterization of
46 Bretherton and Park (2009) determines the cumulus-base mass flux from boundary layer updraft properties
47 rather than ad-hoc closure assumptions typical of AR4 climate models. Using approaches such as these,
48 many climate models simulate boundary-layer cloud radiative properties and vertical structure more
49 accurately than at the time of AR4 (e.g., Köhler et al., 2011; Park and Bretherton, 2009).

50 51 *7.2.3.5 High-Resolution Global Modelling*

52
53 Since AR4, increasing computer power has made it possible to simulate the global circulation of the
54 atmosphere and associated clouds with greater resolution. There have been three types of developments.
55 First, models have been run with resolution that is higher than in the past, but not so high that cumulus
56 clouds can be resolved. Second, models have been run with resolution high enough to resolve (or “permit”)
57 large individual cumulus clouds over the entire globe. In a third approach, the parameterizations of global

1 models have been replaced by embedded cloud-resolving models. The first approach is a continuation of the
2 broad evolution of climate models toward finer simulated scales, so it is assessed in Chapter 9. The other
3 approaches discussed below, are computationally intensive and have only been applied to weather and short
4 climate simulations. Because they overcome many of the parameterization challenges associated with clouds,
5 they make an interesting complement to conventional global atmospheric models.

6 7 *7.2.3.5.1 Global cloud-resolving models*

8 A Japanese global cloud-resolving model (GCRM) called NICAM (Miura et al., 2005; Tomita et al., 2005)
9 has been run with a grid spacing of as little as 3.5 km. At present it can only be used for relatively short
10 simulations of a few simulated weeks or months on the fastest supercomputers. Even with a 3.5 km grid
11 spacing, NICAM can only resolve large cumulus clouds, not the eddies within boundary-layer clouds.
12 Parameterizations of cloud microphysics, radiation, and turbulence are still needed, but these benefit from
13 the fine grid spacing.

14
15 NICAM simulates many features of deep convection that are very challenging for conventional GCMs. Sato
16 et al. (2009) show that NICAM can simulate the diurnal cycles of precipitation associated with land-sea
17 breezes and thermally induced topographic circulations; results improve for finer grid spacing. Oouchi et al.
18 (2009) shows NICAM accurately simulates most features of the Asian summer monsoon. Inoue et al. (2010)
19 showed the cloudiness simulated by NICAM is in good agreement with observations from CloudSat and
20 CALIPSO, but the simulation of cloud ice is not satisfactory and requires an improved parameterization of
21 ice microphysics. Iga et al. (2010) found that changes in the turbulence and cloud microphysics
22 parameterizations of NICAM can strongly affect the upper-level cloudiness, total precipitation, and Hadley
23 circulation. In particular, the Hadley circulation weakens when the simulated high cloud amount increases,
24 due to reduced tropospheric radiative cooling. Even with such fine grid resolution, microphysical
25 parameterization uncertainties can significantly affect simulated climate.

26
27 Within the next decade, it may become computationally feasible to use GCRMs for century-long climate
28 change simulations. Meanwhile, GCRMs can be used in shorter numerical experiments that shed light on
29 results from lower-resolution models.

30 31 *7.2.3.5.2 Models that use embedded cloud-resolving models as “super-parameterizations”*

32 Grabowski and Smolarkiewicz (1999) and Grabowski (2001) pioneered the use of CRMs as ‘super-
33 parameterizations’, i.e., substitutes for the parameterizations in conventional GCMs. Khairoutdinov and
34 Randall (2001) tested the idea in a version of the Community Atmosphere Model (CAM). Tao et al. (2009)
35 developed a similar model using a different global model and different CRM. As with NICAM, cloud
36 microphysics, radiation, and turbulence must still be parameterized; the computational cost is intermediate
37 between GCRMs and conventional GCMs

38
39 The “super-parameterized” CAM gives realistic simulations of the diurnal cycle of precipitation
40 (Khairoutdinov et al., 2005; Pritchard and Somerville, 2010) and the MJO (Benedict and Randall, 2009). Its
41 climatological biases in precipitation and mean circulation are comparable to other climate models, including
42 excessive rainfall in boreal summer over the western Pacific Ocean and southern Asia (Khairoutdinov et al.,
43 2005) and under-prediction of marine stratocumulus clouds (Blossey et al., 2009; Wyant et al., 2009). Stan et
44 al. (2010) coupled the super-parameterized atmosphere model with a global ocean model for 20 simulated
45 years, giving an improved simulation of the Asian summer monsoon (DeMott et al., 2011) and mean rainfall
46 biases compared to the uncoupled model version, and an encouraging simulation of El Niño - Southern
47 Oscillation.

48
49 Super-parameterized global atmospheric models can be used for climate simulations spanning decades or
50 centuries with present-day computers. Like GCRMs, these models give improved simulations of climate
51 variability on a range of time scales, but will benefit from improved parameterizations of turbulence and
52 microphysics.

53 54 *7.2.4 Cloud and Water-Vapour Feedback*

55
56 Climate feedbacks are a central concern for projecting the magnitude of climate change, because they
57 determine the sensitivity of climate to external forcing agents. The overall climate sensitivity of current

1 climate models is assessed in Chapter 9, but the individual contributing feedbacks are assessed in the
2 relevant process-oriented chapters. Water vapour, lapse rate and cloud feedbacks are all assessed in this
3 chapter because they involve moist atmospheric processes closely linked to clouds. In combination they
4 produce most of the simulated climate feedback and also most of its intermodel spread.
5

6 Feedbacks are often expressed as a TOA net downward radiative flux change per degree of global surface
7 temperature increase; in this section they are converted to dimensionless feedback factors by further
8 multiplying by $0.31 \text{ K W}^{-1} \text{ m}^2$ (Roe and Baker, 2007). Such feedback factors from different processes add to
9 give the total feedback factor f . The equilibrium climate sensitivity is inversely proportional to $1-f$, so a total
10 feedback factor of $f=0.6$ (typical of GCMs) would amplify the (blackbody) climate sensitivity by a factor of
11 2.5. Regional feedbacks are sometimes estimated based on surface, rather than TOA, energy fluxes;
12 however, one must then also consider turbulent latent and sensible heat fluxes, complicating the analysis and
13 making it a less useful predictor of the overall coupled system response, so we do not adopt this approach.
14

15 7.2.4.1 *Cloud Altitude Feedback Mechanisms Involving High-Level Clouds*

16
17 High clouds exert little net TOA radiative effect in the current climate due to near-compensation between
18 their longwave and shortwave cloud radiative effects (Kiehl, 1994). Nonetheless, systematic changes in their
19 properties could produce a significant radiative feedback by altering this balance.
20

21 New studies confirm that in typical global warming scenarios, longwave cloud feedback is consistently
22 positive across CMIP3 climate models, with an interquartile range (IQR) of 0.06–0.12 (i.e., $0.2\text{--}0.4 \text{ W m}^{-2}$
23 K^{-1}) (Soden and Vecchi, 2011; Zelinka et al., 2011a), and is primarily due to high clouds (Zelinka et al.,
24 2011a). The dominant driver of longwave cloud feedback appears to be a robust consequence of global
25 warming - an increase in the heights of the tropopause and the main level at which the deepest convective
26 clouds stop rising and cloudy air flows outward, tentatively attributed in AR4 to the so-called fixed anvil-
27 temperature (or FAT) mechanism (Hartmann and Larson, 2002). According to this mechanism, the outflow
28 level from deep convective systems is determined ultimately by the highest point at which water vapour
29 amounts are sufficient to emit significant infrared radiation; this point tends to occur at the same temperature
30 regardless of climate, provided that relative humidity does not change too much, and therefore occurs at a
31 higher altitude in a warmer climate. A positive cloud altitude feedback results because an otherwise identical
32 cloud located higher in the atmosphere exerts a stronger greenhouse effect. New research has confirmed that
33 the hypothesised behaviour is simulated in more realistic models (Kuang and Hartmann, 2007; Kubar et al.,
34 2007), and explains the systematic tendency toward positive cloud feedback in GCMs (Zelinka and
35 Hartmann, 2010). Zelinka et al. (2011b) find that in the CMIP3 models, the IQR of cloud altitude feedback
36 factor is 0.08–0.15, which is even stronger than the overall longwave cloud feedback.
37

38 The observational record allows us to verify various elements of the expected FAT response to global
39 warming. The global tropopause is rising as expected (Chapter 2). Cloud height is correlated to regional and
40 seasonal changes in near-tropopause temperature structure (Eitzen et al., 2009), although the response is
41 affected by changes in stratospheric circulation (Chae and Sherwood, 2010; Eitzen et al., 2009).
42

43 7.2.4.2 *Feedback Mechanisms Involving the Amount of Middle and High Cloud*

44
45 Nearly all GCMs also simulate an overall reduction in middle and high cloud amount in warmer climates,
46 especially in the subtropics (Trenberth and Fasullo 2009; Zelinka and Hartmann 2010). This reduction is
47 geographically correlated with simulated subtropical drying (Meehl et al., 2007), suggesting that it is tied to
48 large-scale circulation changes (Sherwood et al., 2010; Wetherald and Manabe, 1980). The upward mass flux
49 in deep clouds also decreases in a warmer climate (Section 7.2.5.1) which might affect cloud cover in ways
50 difficult to capture in current models. In global average, both middle and high-level simulated cloudiness
51 reductions cause a positive shortwave feedback, but the high cloud reductions also compensate nearly half of
52 the longwave cloud altitude feedback. This may explain why researchers did not identify the important role
53 of cloud altitude feedbacks sooner. From Figure 8 of Zelinka and Hartmann (2011b) the IQR of the CMIP3
54 multimodel mean net cloud feedback factor from middle and high clouds is estimated to be 0.08–0.16. This
55 is similar to the cloud altitude feedback alone, so we conclude that the additional net feedback due to
56 mid/high cloud amount reductions is small.
57

1 Bender et al. (2011) find decreasing trends in satellite-derived subtropical cloud cover, but also note large
2 uncertainties involved in correcting for satellite radiometer and orbital drift as well as cross-calibration
3 between satellites (see Chapter 2). Tselioudis and Rossow (2006) predict reduced storm-track cloud cover in
4 warmer climates based on observed present-day relationships with meteorological variables combined with
5 model-simulated changes to those driving variables. In agreement with the above analysis, they found that
6 this reduction contributed little to net cloud feedback, due to compensating longwave and shortwave
7 contributions.

8
9 Thin cirrus clouds exert a net warming effect on climate, and cover a significant area. These clouds could
10 therefore exert an important feedback on climate if their area changed relatively moderately with global
11 temperature (e.g., Rondanelli and Lindzen, 2010). We find no compelling evidence from observations,
12 process models, or GCMs suggesting such a feedback is important, and the CMIP3 multimodel mean change
13 of thin high cloud fraction is smaller than for other cloud types (Zelinka et al., 2011a). A caveat is that these
14 clouds are challenging to accurately simulate in climate models (see Chapter 9).

15
16 A positive contribution to net cloud feedback also comes from the anticipated poleward shift of storm tracks
17 in a warmer climate. Even if storm track clouds remained unaltered, this shifts clouds to latitudes of weaker
18 sunlight, decreasing the planetary albedo. Such a shift occurs in most models (Yin, 2005) and has appeared
19 in recent cloud and other data (see Chapter 2); similar shifts are also seen in indicators of the edge of the
20 tropics (Scheff and Frierson, 2011). The impact of the observed shift on clouds, particularly the reduction in
21 subtropical cloud cover, appears to be significant and would imply a strong positive feedback if it were due
22 to global warming (Bender et al., 2011). Recent studies call into question how much of the observed shifts
23 are temperature-driven vs. ozone-driven [Chapter 10.3.x], so the true magnitude of this feedback
24 contribution remains highly uncertain. As most GCMs produce too little storm-track cloud in the southern
25 hemisphere, it has been suggested that they underestimate this feedback (Trenberth and Fasullo, 2010) even
26 if the model-predicted shifts are correct.

27 28 *7.2.4.3 Feedback Mechanisms Involving Low Clouds*

29
30 Low clouds exert a strong net cooling effect on the Earth, such that if their coverage or water content were
31 climate-sensitive a feedback would result. Feedback contributions from low clouds continue to differ
32 significantly among models, and to cause most of the spread in global climate sensitivity among GCMs for
33 both transient and equilibrium simulations (e.g., Dufresne and Bony, 2008). Zelinka et al. (2011a) show that
34 the CMIP3 IQR for low cloud feedback factor is 0.09–0.15; all analysed models had positive low cloud
35 feedback, but of widely differing magnitudes. The feedback derives from slight decreases in low cloud
36 cover, which in multimodel mean occurs throughout low and mid-latitudes (Zelinka et al., 2011b), though
37 the geographical pattern varies widely between models (Webb et al., 2012).

38
39 No robust feedback mechanisms involving tropical and mid-latitude low clouds have yet been established,
40 though many possible mechanisms for both positive and negative feedbacks have been proposed. It has long
41 been suggested that cloud water content could increase in a warmer climate simply due to the higher water
42 vapour mixing ratio (WVMR) in sub-cloud air or more condensation per unit height in an adiabatic cloudy
43 updrafts, but this argument ignores the physics of crucial cloud-regulating processes like precipitation
44 formation and turbulence. Observational evidence discounting the suggested effects was reported in AR4.

45
46 As noted in Section 7.2.1.3.2, at each latitude, low cloud cover tends to favor the coldest oceans, which can
47 naively be interpreted to imply a positive feedback. This relationship is thought to be regulated by lower-
48 tropospheric stability, but different measures of stability that are similarly skilful in the current climate imply
49 significantly different cloud changes in a warmer climate (Section 7.2.1.3). Likewise, interannual variations
50 of ocean surface temperature in marine stratocumulus regions tend to anti-correlate with changes in cloud
51 cover and water content, again suggesting a negative feedback if taken at face value (Eitzen et al., 2011).
52 However, since these regional variations are accompanied by local atmospheric stability changes that would
53 differ from those accompanying a global warming, this interannual relationship is unlikely to indicate the
54 global feedback (see Section 7.2.4.3.4).

55
56 Studies since the AR4 have yielded some further insight into the diversity of low-cloud feedbacks in GCMs.
57 Several climate models that have been run with land removed (a so called aquaplanet) show cloud feedback

1 comparable to their usual configuration, suggesting that neither land-surface feedbacks nor land-induced
2 heterogeneity is crucial in determining the low-cloud response to warming in the full GCMs (Medeiros et al.,
3 2008). Low-cloud feedback in GCMs arises from a mix of cumulus and stratocumulus cloud feedbacks
4 (Williams and Tselioudis, 2007; Williams and Webb, 2009; Xu et al., 2010). Bony and Dufresne (2005)
5 found that that the intermodel spread among tropical feedbacks derives mainly from the regimes of moderate
6 subsidence that support both these cloud types. Webb et al. (2012) found that the feedback factor and its
7 spread is largest in cool-ocean (stratocumulus) regimes and shallow cumulus regimes covering a larger
8 fraction of the subtropics.

9
10 Other post-AR4 studies have probed how different parameterizations affect simulated low-cloud feedbacks.
11 For example, an increased positive cloud feedback in two successive versions of the NCAR GCM is due
12 mainly to changes in the representation of shallow cumulus convection, but with significant compensation
13 from the boundary-layer parameterisation, and the biggest simulated cloudiness reductions occur in the
14 Southern Ocean stratocumulus regime due to interactions between cumulus and stratocumulus (Gettelman et
15 al., 2011b). A single-column diagnostic study of Zhang and Bretherton (2008) found that the interaction
16 between boundary layer, shallow and deep convective parameterisations in transporting moisture upward to
17 form clouds was crucial in determining the cloud albedo feedback, even in a stratocumulus regime.

18
19 Cloud feedbacks have been examined in first-generation global cloud-resolving models and
20 “superparameterized” GCMs, (Section 7.2.3.5), which avoid cumulus parameterization. These models have
21 shown increases in low-cloud cover in warmer climates (Wyant et al., 2009; Wyant et al., 2006), predicting
22 climate sensitivities at or below the low end from GCMs. GCMs with superparameterisations do not,
23 however, simulate low clouds any better than (or even as well as) traditional GCMs, apparently due to the
24 very fine grid (~100 m or better) required for proper, explicit simulation of the cloudy boundary layer
25 (Section 7.2.2.1, Blossey et al., 2009). Limited-area high-resolutions models have also been used to estimate
26 cloud feedbacks (Lauer et al., 2010; Xu et al., 2010), and have yielded a range of estimates even broader than
27 those of global models. These models and methodologies are less well tested than traditional GCMs, so we
28 assign relatively little confidence to their feedback behaviour at this time.

30 7.2.4.4 *Feedbacks Involving Changes in Cloud Optical Depth*

31
32 Another possible cloud feedback mechanism involves changes in cloud phase. At mixed-phase temperatures
33 of –40 to 0°C, cloud ice particles that contribute most to light scattering are typically several-fold larger than
34 cloud water drops (e.g., Mitchell et al., 2010), so a given mass of cloud ice reflects less sunlight than the
35 same mass of cloud water droplets in today’s atmosphere. As climate warms, the shift from ice to liquid
36 clouds would raise albedos if cloud water mass did not change. The resulting negative cloud feedback
37 appears in GCMs (Senior and Mitchell, 1993), with a magnitude that correlates with the simulated amount of
38 cloud ice in mixed phase clouds (Tsushima et al., 2006). It is unlikely however that cloud water content
39 would remain fixed as clouds changed phase, and the key physics is not represented in climate models, so
40 this feedback mechanism is highly uncertain.

41
42 Zelinka et al. (2011b) isolated the component of simulated cloud feedback due to changes in cloud optical
43 depth in a set of CMIP3 GCMs. The global-mean net feedback scattered around zero, but, there was a
44 tendency toward slightly reduced optical depths at low and middle latitudes, and increases at latitudes
45 poleward of 50° yielding a negative local feedback that they attributed partly to phase changes and partly to
46 the greater cloud water amounts expected from increased poleward moisture transport (Vavrus et al., 2009).

48 7.2.4.5 *Feedback from Arctic Cloud Interactions with Sea Ice*

49
50 Arctic clouds, despite their low altitude, have a net warming effect at the surface in the present climate
51 because their downward emission of infrared radiation over the year outweighs their reflection of sunlight
52 during the short summer season. However, they also cool the atmosphere, so their effect on the energy
53 balance of the whole system is ambiguous and depends on the details of the vertical cloud distribution and
54 the impact of cloud radiative interactions on ice cover.

55
56 Visual cloud reports (Eastman and Warren, 2010) and lidar observations available since AR4 (Kay and
57 Gettelman, 2009; Palm et al., 2010) now agree that low cloud cover over Arctic oceans is inversely

1 correlated to sea ice amount, with open water producing more cloud. The observed effect is weak in boreal
2 summer, when the melting sea-ice is at a similar temperature to open water and stable boundary layers with
3 extensive low cloud are common over both surfaces, and strongest in boreal autumn when cold air flowing
4 over regions of open water stimulates cloud formation by boundary-layer convection (Kay and Gettelman,
5 2009; Vavrus et al., 2011). Kay et al. (2011) show that a GCM can represent this seasonal sensitivity of low
6 cloud to open water, but only with an appropriate boundary-layer cloud parameterization. Vavrus et al.
7 (2009) show that in a global warming scenario, GCMs simulate more Arctic low-level cloud in all seasons,
8 but especially during autumn and winter when open water and very thin sea ice increase considerably,
9 decreasing low-level stability and increasing upward moisture transport from the surface to levels at which it
10 forms clouds.

11
12 A negative Arctic cloud feedback was suggested by Liu et al. (2008) on the basis that observed surface
13 warming in recent decades was greater under clear-sky than under cloudy conditions, but this argument was
14 not tested in a climate model and does not control for the large correlated effects of weather variability on
15 both clouds and surface temperature. Gagen et al. (2011) present tree-ring evidence that summertime Arctic
16 cloud cover was negatively correlated with Arctic temperatures over the last millennium, which is consistent
17 with the conclusions of the above studies assuming there was less ice during warmer periods. While Gagen
18 et al. (2011) presented this as evidence of negative cloud feedback, they ignored the year-round greenhouse
19 effect of clouds, which could change the imputed feedback to a positive one (Palm et al., 2010). Note that
20 there are pitfalls to using natural climate variations to infer cloud feedbacks, described in Section 7.2.4.3.7,
21 that apply here as well.

22 23 *7.2.4.6 Fast Adjustment of Clouds and Precipitation to a CO₂ Change*

24
25 Climate feedbacks are usually estimated by comparing steady-state simulations with control and doubled
26 CO₂, and analysing the contribution of different processes to the simulated changes in radiation balance,
27 normalized by the change in global-mean surface air temperature (e.g., Soden and Held, 2006).

28
29 Gregory and Webb (2008) partitioned the transient response of the radiation balance of GCMs to an
30 instantaneous doubling of CO₂ into a ‘fast’ (sub-seasonal) adjustment in which the land surface, atmospheric
31 circulations and clouds respond to the radiative effect of the CO₂ increase, and an ‘SST-mediated’ response
32 that develops more slowly as the oceans warm. They found that in some climate models, fast adjustment of
33 clouds can have comparable top-of-atmosphere radiative effects to the ensuing SST-mediated cloud changes.
34 However, Andrews and Forster (2008) found that this behavior was exceptional and that on average, fast
35 cloud adjustments in a suite of climate models causes less than 20% of their equilibrium radiative feedback;
36 low cloud adjustments have the biggest global net radiative impact (Colman and McAvaney, 2011). Fast
37 adjustment may cause clouds to respond slightly differently during a transient climate change (in which SST
38 changes have not caught up to CO₂ changes) than after equilibrium is reached and SST changes have been
39 fully expressed. Studies of fast adjustment of climate models to a step CO₂ increase have also shown reduced
40 global-mean precipitation (Cao et al., 2011) and cloud cover (Andrews and Forster, 2008), a shift of tropical
41 clouds and precipitation from ocean to land (Lambert et al., 2011; Wyant et al., 2011), and shallowing of
42 marine stratocumulus cloud (Caldwell and Bretherton, 2009; Wyant et al., 2011). The ensuing response to
43 warming SST reverses many of these trends, increasing global-mean precipitation and shifting precipitation
44 and high clouds back over the tropical oceans (Cao et al., 2011).

45 46 *7.2.4.7 Observational Constraints on Global Cloud Feedback*

47
48 A number of studies since AR4 have attempted to constrain cloud feedback (or total climate sensitivity) from
49 observations; here we discuss those using modern cloud, radiation or other measurements. Section [12.5]
50 discusses those based on past temperature data and forcing proxies.

51
52 One approach is to seek observable aspects of present-day cloud behaviour that reveal cloud feedback. In at
53 least two climate models, large sets of runs with nonstandard parameter settings produce feedback strengths
54 that correlate with the amount or water content of cloud simulated for the present day (Williams and Webb,
55 2009), but in other models this does not happen (Yokohata et al., 2010), and the resulting relationships do
56 not hold across multiple models e.g., CMIP3 (Gettelman et al., 2011a). Among the AR4 models, net cloud
57 feedback is strongly correlated with mid-latitude relative humidity (Volodin, 2008) and with characteristics

1 of the southern-hemisphere storm track (Trenberth and Fasullo, 2010); if valid either regression relation
2 would imply a relatively strong positive cloud feedback in reality, but no mechanism has been proposed to
3 explain or validate these empirical relationships. Likewise, Clement et al. (2009) found realistic decadal
4 variations of low cloud over the North Pacific in only one model (HadCM3) and argued that the relatively
5 strong cloud feedback in this model should therefore be regarded as more likely, but provided no evidence
6 for such a link. Chang and Coakley (2007) examined midlatitude maritime clouds and found cloud thinning
7 with increasing temperature, consistent with a positive feedback, while Gordon and Norris (2010) found the
8 opposite result in following a methodology that tried to isolate thermal and advective effects. In summary,
9 there is no evidence of a robust link between any of the noted observables and the global feedback, though
10 some apparent connexions are tantalising and are being further studied.

11
12 Several studies have attempted to derive long-term climate sensitivity from interannual relationships between
13 global-mean observations of top-of-atmosphere radiation and surface temperature. One problem with this is
14 the different spatial character of interannual and long-term warmings; another is that the methodology can be
15 confounded by cloud variations not caused by those of surface temperature (Spencer and Braswell, 2008). A
16 range of climate sensitivities has been inferred based on such analyses (Forster and Gregory 2006; Lindzen
17 and Choi, 2011). Crucially, however, among different GCMs there is no correlation between the interannual
18 and long-term cloud-temperature relationships (Dessler, 2010), contradicting the basic assumption of these
19 methods. On the other hand the GCMs, on average, do predict a cloud radiative response to these high-
20 frequency global temperature changes that is consistent with the global feedback response and with
21 observations, increasing the credibility of their predictions at longer timescales (Dessler, 2010). More
22 recently there is interest in relating the time-lagged correlations of cloud and temperature to feedback
23 processes (Spencer and Braswell, 2010) but again these relationships appear to reveal only a model's ability
24 to simulate ENSO or other modes of interannual variability properly, which cannot be translated directly into
25 cloud feedback on long-term global warming (Dessler, 2011).

26
27 While a number of studies have proposed methods to infer the long-term cloud feedback from observed
28 variability, for a method to be accepted it should have a sound physical basis and be shown to work
29 consistently when applied to climate models. No method yet proposed passes both tests. Moreover, some
30 model studies show that the response of global cloud radiative effect to a global warming is sensitive to
31 relatively subtle details in the geographic warming pattern, such as the slight hemispheric asymmetry due to
32 the lag of southern ocean warming relative to northern latitudes (Senior and Mitchell, 2000; Yokohata et al.,
33 2008). Cloud responses to specified uniform ocean warming without CO₂ increases are not the same as those
34 to CO₂-induced global warming simulated with more realistic oceans (Ringer et al., 2006), partly because of
35 fast adjustment (Section 7.2.4.3.6) and because low clouds also feed back tightly to the underlying surface
36 (Caldwell and Bretherton, 2009). Simulated cloud feedbacks also differ significantly between colder and
37 warmer climates in some models (Crucifix, 2006; Yoshimori et al., 2009). These sensitivities highlight the
38 challenges facing any attempt to infer long-term cloud feedbacks from simple data analyses.

40 [INSERT FIGURE 7.5 HERE]

41 **Figure 7.5:** CFMIP figure on cloud feedbacks in CMIP5 models. [PLACEHOLDER FOR SECOND ORDER DRAFT:
42 CMIP3 version used as placeholder.]

44 7.2.4.8 Feedback Synthesis

45
46 Together, water vapour-lapse rate and cloud feedback are the principal determinants of climate sensitivity.
47 The combined global water vapour-lapse rate feedback simulated by all GCMs is strongly positive (feedback
48 factor IQR 0.28–0.33), similar to AR4, and has the magnitude expected from simple physical arguments that
49 on global scales, absolute humidity changes much more than relative humidity in a perturbed climate.

50
51 Cloud feedbacks on CO₂-induced climate change are less certain, but the combined evidence suggests a
52 substantial positive net feedback from clouds. Two estimates of the IQR of overall cloud feedback factor,
53 based on two slightly different groups of CMIP3 GCM simulations are 0.11–0.33 (Table 1 of Soden and
54 Held, 2006), and 0.17–0.28 (Figure 8 of Zelinka et al., 2011a). Differences between models have not reduced
55 since AR4 [TBC] and dominate the spread in model climate sensitivities. However, new approaches to
56 diagnosing cloud feedback in GCMs have clarified robust cloud responses, while continuing to implicate low
57 cloud cover as the most important source of intermodel spread in simulated cloud feedbacks. Some new

1 approaches to observation and mechanistic understanding of cloud feedback have been taken, using a
2 hierarchy of models and observations on local and global scales.

3
4 Several physical processes contribute to a positive net cloud feedback, as summarized in Figure 7.6. First,
5 high clouds to rise in altitude and thereby exert a stronger greenhouse effect in warmer climates. This
6 altitude feedback mechanism is well understood, has theoretical and observational support, occurs
7 consistently in GCMs and other models, and explains about half of the multimodel-mean positive cloud
8 feedback. Second, middle and high level cloud cover tends to decrease in warmer climates even within the
9 storm tracks and ITCZ, consistent with the reduction of upward, cloudy air fluxes required to balance the
10 hydrological cycle in an atmosphere with more water vapour. This decreases the albedo more than the
11 greenhouse effect, adding positive feedback. Third, observations and most models suggest storm tracks shift
12 poleward in a warmer climate, drying the subtropics and moistening the high latitudes, which causes further
13 positive feedback via a net shift of cloud cover to latitudes that receive less sunshine. Most GCMs also
14 predict low cloud amount decreases especially in the subtropics, another source of positive feedback, though
15 there is wide spread in this feedback and it lacks a well-accepted theoretical basis. Over high latitudes,
16 models suggest warming-induced transitions from ice to water clouds may cause clouds to become more
17 reflective, but this optical depth feedback operates over too small a region to be globally significant. While
18 other feedbacks remain possible and uncertainties remain –particularly for low cloud amount– there is no
19 consistent observational evidence, consensus among models, nor robust physical argument that would favour
20 these having a negative feedback contribution over a positive one.

21
22 Since all established globally-significant feedback mechanisms are positive, a positive overall cloud
23 feedback is more likely than a negative one. How much more likely depends on the likely strength of any
24 other mechanisms that may exist. Our chief guidance on this question is from GCMs. As of yet, the wide
25 variety of formulations and resulting behaviour in these models has not produced a single example [TBC]
26 where other mechanisms have been strong enough to outweigh unequivocally the known sources of positive
27 feedback. In the absence of supporting evidence, we judge that cloud feedback outside the range of any
28 current CMIP3 GCM is unlikely (less than 33% probability), and that such cloud feedback is no more likely
29 to lie below the CMIP3 range than above it. The lowest cloud feedback for any CMIP3 model was slightly
30 positive (Soden and Held, 2006), so this reasoning implies that the net cloud feedback is likely (83% chance)
31 positive. Note that this conclusion is independent of constraints on climate sensitivity from observed trends
32 or palaeoclimate information as discussed in Section 12.4.

33 [INSERT FIGURE 7.6 HERE]

34 **Figure 7.6:** Robust cloud responses to greenhouse warming simulated by the CMIP3 multimodel ensemble. Panel (a) is
35 a schematic latitude-altitude section showing typical cloud types in a pre-industrial climate. Grey (white) indicates
36 clouds composed predominantly of liquid water (ice). Raindrops and snowflakes indicate the typical precipitation type.
37 Dotted line indicates the typical freezing level, and purple dashed line indicates the tropopause. Panel (b) shows the
38 same cross section for a warmer climate, with arrows denoting the movement of different boundaries. Tropical deep
39 convection regions narrow and intensify, the subsidence regions of the subtropics widen poleward, with most GCMs
40 projecting low cloud decreases in this area, and storm track cloud and precipitation also shift poleward. Cirrus cloud
41 tops rise in lockstep with the tropopause, helping induce positive longwave cloud feedbacks. The rising freezing level
42 causes more cloud to become liquid, contributing to increased optical thickness of high latitude clouds in the CMIP3
43 multimodel mean. [PLACHOLDER FOR SECOND ORDER DRAFT: CMIP5]

44 7.2.5 Basis of Precipitation Changes in Cloud Physical Processes

45
46 This section reviews fundamental process-level knowledge relevant to changes of precipitation
47 characteristics in warmer climates and their relation to clouds. Observed trends in mean and extreme
48 precipitation are discussed in Chapters 2 and 10; changes in hurricanes and other dynamical phenomena, and
49 precipitation projections for specific regions, are discussed in Chapters 11, 12 and 14.

50 7.2.5.1 Coupling of Large-Scale Trends in Clouds and the Hydrological Cycle

51
52 In a perturbed climate, regional changes in cloud cover and type will inevitably accompany regional changes
53 in the hydrological cycle. The CMIP3 coupled climate models as a group predict that in 21st century global
54 warming scenarios, the ITCZ will narrow and intensify, the subtropics will dry, and the storm tracks will
55 move poleward (Held and Soden, 2006; Meehl et al., 2007); helping induce the cloud responses discussed in

1 Section 7.2.4 and shown in Figure 7.6. The strong geographical relation between projected precipitation and
2 cloud changes can be seen in CMIP multimodel mean results (IPCC 2007) [PLACEHOLDER FOR
3 SECOND ORDER DRAFT: CMIP5 updates].
4

5 Within the tropics, individual climate models simulate regional changes in the zonally asymmetric part of
6 mean rainfall patterns and circulation as the climate warms. The detailed geographical pattern of these
7 changes is model-dependent (Bony et al., 2004; Neelin et al., 2006), and they lead to annual-mean deep
8 convective cloud changes in each model that are quite similar to the corresponding changes in mid-
9 tropospheric vertical motion (Bony et al., 2004), such that up to 70% of the longwave and shortwave cloud
10 radiative forcing changes at a typical location over the tropical oceans can be ascribed to changes in the
11 vertical motion at that location (Wyant et al., 2006). This point motivates such analysis techniques as
12 vertical-velocity binning (Bony et al., 2004) for separating local dynamically driven cloud changes from the
13 residual ‘thermodynamic’ cloud changes. Over the scale of the tropics or the entire globe, regions of
14 enhanced mean ascent balance those of more mean descent, so these dynamical effects on cloud response
15 largely cancel while the thermodynamic cloud changes need not (Bony et al., 2004).
16

17 7.2.5.2 *Changes in Precipitation Extremes*

18
19 Studies since AR4 have focused new attention on the climate-dependence of rainfall extremes down to
20 hourly time scales. The expectation has been that extreme precipitation is limited by the precipitable water
21 content of the atmosphere in the storm environment, which increases at roughly 7% per °C of warming
22 (“Clausius-Clapeyron” or CC scaling) if relative humidity is constant. However, not only can relative
23 humidity vary significantly, but intensified latent heating could feed back on cloud-scale dynamics so as to
24 alter the efficiency with which precipitable water is condensed (O’Gorman and Schneider, 2009); such
25 changes are sensitive to factors other than surface temperature (Berg et al., 2009).
26

27 Lenderink and van Meijgaard (2008, 2010) report that extremes of daily rain accumulation at several sites in
28 Europe show approximate CC scaling with respect to weather variations (not temperature trends), but
29 summertime extremes at the hourly time scale showed a scaling twice this strong, probably due to stronger
30 convective storms on hotter days. Lenderink and van Meijgaard (2010) link this to atmospheric humidity,
31 finding an even stronger supersensitivity of 17% per °C of dewpoint. These results were roughly reproduced
32 by regional model simulations. Similar supersensitivities appear in maritime tropical rainfall rates (Allan and
33 Soden, 2008; Allan et al., 2010). However, they do not appear at most sites in the U.S. (Shaw et al., 2011) or
34 Australia (Jones et al., 2010b), where hourly rain scaling is more erratic but close to CC on average. In some
35 datasets, particularly the Australian data, scaling turns negative at the highest temperatures which are
36 associated with dry conditions.
37

38 The interpretation of these results is not straightforward, especially since weather relationships cannot be
39 assumed to apply directly to climate changes. Berg et al. (2009) point out that the temperature scaling
40 inferred from the European data can weaken significantly if one controls for other factors such as storm type.
41 Allan et al. (2010) note that modelled and observed trends in maritime convective extremes since the 1980’s
42 are both close to CC, but that weather-related fluctuations show scaling several times stronger; this reflects
43 the relatively strong sensitivity of tropical convection to local maxima of temperature as opposed to overall
44 means (Johnson and Xie, 2010). Idealised calculations using regional (Muller et al., 2011) or cloud-resolving
45 (Romps, 2011) models show extremes scaling with or somewhat below CC in idealised global warming
46 scenarios, possibly because humidity increases more slowly near the surface than aloft.
47

48 Regional models are inconsistent in predicting how rainfall extremes will change with global warming,
49 ranging from CC scaling to nearly double this; predicted changes to extremes are also regionally diverse,
50 exceeding CC by factors of several in regions that receive more rain but decreasing in those receiving less
51 (Lenderink and van Meijgaard, 2010). This makes generalised statements difficult. However, all results point
52 to greater rainfall extremes in warmer climates except in places of significant rainfall decline. The best guess
53 as to the magnitude of the increases for typical conditions (e.g., where rain climatology matches the global
54 mean) is probably close to 7% per °C, but this remains uncertain especially for extremes on sub-daily time
55 scales.
56

7.2.6 Anthropogenic Sources of Cloudiness

Human activity can be a source of additional cloudiness through specific processes involving a source of water vapour in the atmosphere. We discuss here the impact of aviation and irrigation on water vapour and cloudiness. The impact of water vapour sources from fossil fuel combustion at the Earth's surface is thought to be negligible.

7.2.6.1 Contrails and Contrail-Induced Cirrus

Aviation jet engines emit hot moist air which can form line shaped persistent condensation trails (contrails) under certain atmospheric conditions. These have been observed to spread into large cirrus sheets which may persist for several hours, and observational studies confirm their overall positive net radiative forcing impact (Haywood et al., 2009). Aerosol emitted within the aircraft exhaust can also affect cloudiness. This last effect is classified as an aerosol-cloud interaction and is treated as part of Section 7.4.

Persistent contrails can form in the upper troposphere in air that is already supersaturated with respect to ice and colder than -40°C . They are composed of ice crystals that are typically smaller than background cirrus (Frömming et al., 2011; Heymsfield et al., 2010), they trap longwave radiation and reflect solar radiation leading to a pronounced diurnal cycle in radiative forcing (Burkhardt and Kärcher 2011; Rap et al., 2010b; Stuber and Forster, 2007).

Forster et al. (2007) estimated the 1750–2005 radiative forcing from persistent linear contrails as 0.01 W m^{-2} (0.003 to 0.03 W m^{-2} 90% uncertainty range). They also estimated a range of forcing from 1750 to 2000 for aviation induced cirrus of 0.03 W m^{-2} (with a 0.01 to 0.08 W m^{-2} 90% uncertainty range). Lee et al. (2009) scaled the IPCC AR4 estimates of the persistent contrail RF to account for revised fuel use estimates, propulsive efficiency and flight routes, which resulted in a 18% increase for 2005. Rap et al., (2010b) found that contrails preferentially formed in conditions where there was already considerable cloud present, which contributed to lessen their radiative forcing. Kärcher et al. (2010) corrected previous estimates for inconsistencies between contrail cover and averaged optical depth and came up with a range of 0.008 to 0.020 W m^{-2} for the reference year 2000. Overall we adopt a RF estimate of $0.02 \pm 0.01 \text{ W m}^{-2}$ (90% uncertainty range) for persistent linear contrails for the period 1750–2010.

Rap et al. (2010a) strengthened the assessment that aviation contrails are very unlikely at current levels of coverage to have an observable effect on surface temperature and diurnal temperature range. They also found a very small efficacy for the contrail radiative forcing of 31%, smaller than the 59% found in the ECHAM4 model (Ponater et al., 2005).

Contrail induced cirrus forcing estimates have been based on correlating observations (e.g., Boucher, 1999) and may have included cirrus changes that were not directly caused by aviation. Burkhardt and Kärcher (2011) have removed the need for such gross assumptions by modelling the global impact of both line-shaped and spreading contrails within a climate model (Burkhardt and Kärcher, 2009). They estimated a RF of 0.037 W m^{-2} for contrails and contrail cirrus but did not fully quantify the uncertainty. They also found the contrails to reduce the background cirrus cloudiness in the main traffic areas and suggest this gives a negative forcing of roughly -0.007 W m^{-2} . Compounding their assessed errors from spreading rate, optical depth, ice particle shape and radiative transfer would give an error estimate of 67% in agreement with Markowicz and Witek (2011). As this study is a major step forward from previous work we employ their estimate for this assessment, and assess a combined contrail and contrail-induced cirrus AF to be 0.03 W m^{-2} with a 90% uncertainty range of 0.01 to 0.06 W m^{-2} . The upper bound for this forcing mechanism is not well constrained however.

7.2.6.2 Irrigation-Induced Cloudiness

Boucher et al. (2004) estimated a RF due to water vapour from irrigation in the range of 0.03 to 0.10 W m^{-2} but the net climate effect was dominated by the evaporative cooling at the surface and by atmospheric thermal responses to low-level humidification. Regional surface cooling was confirmed by a number of more recent regional and global studies (Kueppers et al., 2007; Lobell et al., 2009). It was also found that the resulting increase in water vapour could induce a small enhancement in precipitation downwind of the major

1 irrigation areas (Puma and Cook, 2010), as well as some regional circulation patterns (Kueppers et al., 2007).
2 Sacks et al. (2009) reported a 0.001 increase in cloud fraction over land (0.002 over irrigated land). This
3 points to an AF no more negative than -0.1 W m^{-2} .

4 5 **7.3 Aerosols**

6 7 **7.3.1 Introduction**

8
9 This section assesses the role of aerosols in the current climate system, focusing on anthropogenic changes in
10 aerosols and their effects. In particular it covers the direct radiative forcing of aerosols, their effects on
11 atmospheric heating and snow/ice surfaces, as well as Earth system feedbacks involving natural and
12 anthropogenic aerosols. Cloud microphysical effects of aerosols are discussed in Section 7.4 although some
13 of the relevant properties of aerosols, such as cloud condensation nuclei (CCN) and ice nuclei (IN), are
14 documented here. The time evolution of aerosols and their forcing are discussed in Chapters 2 and 8, with
15 Chapter 8 also covering changes in natural volcanic aerosols.

16
17 The Section covers material previously assessed in the AR4 of the IPCC (IPCC, 2007). Chapter 2 (Forster et
18 al., 2007) assessed the total direct RF from aerosol to be $-0.5 \pm 0.4 \text{ W m}^{-2}$ and broke this down into
19 components associated with several species. Land albedo changes associated with black carbon (BC) on
20 snow were assessed to be $+0.1 \pm 0.1 \text{ W m}^{-2}$. The semi-direct effect and its contribution to the AF were
21 discussed but not explicitly quantified in Chapter 7 (Denman et al., 2007). Chapter 7 also gave a preliminary
22 assessment of possible earth system feedbacks involving aerosols. The RF uncertainty estimate in AR4 was
23 based on the range of model results and remote observations.

24
25 Since AR4 in-situ and remote observations of aerosol have improved and global aerosol models have
26 become considerably more complex and evaluated against observations. Earth system models are continuing
27 to be developed and some have been used in CMIP5, which allows the investigation of possible
28 biogeochemical aerosol feedbacks on climate. To help improve the forward assessment of uncertainty in
29 aerosol forcing this chapter discusses aerosol science in more detail than AR4, from both an observational
30 and a modelling perspective.

31
32 Radiative forcing estimates rely on knowledge of aerosol emissions and aerosol properties. Aerosol sources
33 and properties are discussed in Section 7.3.2 while Section 7.3.3 assesses the key climate-relevant aerosol
34 properties and Section 7.3.4 discusses aerosol distributions. Section 7.3.5 covers the direct and semi-direct
35 effects of aerosols, which provides the basis for evaluating the anthropogenic component to that effect, in
36 terms of RF and AF; this section also assesses the forcing contribution that results from aerosol-induced
37 changes to the surface properties of snow and ice. Finally Section 7.3.6 updates our understanding of
38 potential aerosol-climate feedbacks.

39 40 **7.3.2 Aerosol Sources and Processes**

41
42 Atmospheric aerosol particles, whether natural or anthropogenic, originate from two distinctively different
43 pathways: direct emissions of primary particles and secondary aerosol formation from gaseous precursors
44 (Figure 7.7). Secondary aerosol formation is initiated by gas phase chemistry that produces a large number of
45 organic and inorganic compounds of different level of oxidation and volatility. Some of these compounds
46 form secondary particulate matter by condensing onto or reacting with pre-existing aerosol particles or cloud
47 droplets. A small fraction of gaseous compounds is capable of producing new aerosol particles by
48 nucleation. Both primary and secondary particles grow in size in the atmosphere by condensation,
49 coagulation and cloud processing. These processes also affect the aerosol chemical composition, size, shape
50 and mixing state. Aerosols are removed from the atmosphere through dry deposition at the surface and wet
51 deposition (including in-cloud and below-cloud rainout).

52 53 **[INSERT FIGURE 7.7 HERE]**

54 **Figure 7.7:** Overview of atmospheric aerosol processes and meteorological variables influencing the aerosol semi-
55 direct, direct and indirect aerosol effects. Red designates gas phase processes and variables; blue designates particulate
56 (aerosol) phase processes and variables; processes and variables relevant to the aerosol direct and semi-direct effects
57 appear in black, while those relevant to the aerosol indirect effects appear in green.

1
2 The main chemical constituents of the atmospheric aerosol are inorganic species (such as sulphate, nitrate,
3 sea-salt), organic species (also termed organic carbon or organic aerosol), black carbon (BC), and mineral
4 species (mostly desert dust). BC, sea salt and dust are introduced into the atmosphere as primary particles,
5 whereas sulphate and nitrate are formed almost entirely in the atmosphere by secondary aerosol formation
6 processes. Organic aerosol (OA) has both primary (POA) and secondary (SOA) sources. The majority of BC,
7 sulphate and nitrate come from anthropogenic sources, whereas sea salt and most of dust is of natural origin.
8 Atmospheric POA is likely to be dominated by anthropogenic sources, whereas SOA is to a larger extent of
9 natural origin (Farina et al., 2010; Hallquist et al., 2009; Kroll and Seinfeld, 2008). Despite earlier
10 recognition of their climatic importance (Adams et al., 2001), it is only recent that nitrate aerosols are being
11 considered in a wider set of models. Emissions of aerosols and aerosol precursors are summarized in Table
12 7.1.

13
14 **[INSERT TABLE 7.1 HERE]**

15 **Table 7.1:** Global and regional anthropogenic emissions important for aerosol formation and tropospheric chemistry.
16 The maximum and minimum values from available inventories are presented. Units for NO_x are Tg NO yr⁻¹, other units
17 are Tg yr⁻¹. Adapted from Granier et al. (2011).

18
19 The climate effects of atmospheric aerosol particles depend on their atmospheric distribution, along with
20 their optical properties and ability to act as CCN or IN. Key quantities for aerosol optical and cloud forming
21 properties are the particle number size distribution, chemical composition, mixing state and shape. These
22 properties are determined by a complex interplay between their sources, atmospheric transformation
23 processes and their removal from the atmosphere (Figure 7.7).

24
25 Sea salt particles are produced at the sea surface by breaking waves (de Leeuw et al., 2011). Emissions of sea
26 salt particles depend mainly on the surface wind speed, and to a lesser extent on the temperature and
27 composition of the sea water; however the effective flux in the atmosphere also depends on atmospheric
28 stability. Since AR4, substantial progress has been made in constraining the total mass, number size
29 distribution and chemical composition of emitted sea salt particles (Evan et al., 2008; Fuentes et al., 2011;
30 Keene et al., 2007; Ovadnevaite et al., 2011). A major new observation has been the frequent presence of
31 organic material in submicron sea salt particles, especially at smallest particle sizes. Process-based estimates
32 of sea salt emissions continue nevertheless to have a significant uncertainty (de Leeuw et al., 2011). These
33 uncertainties in the chemical composition and size of emitted sea-salt particles translate into a large
34 uncertainty on the natural level of CCN in the marine atmosphere, which unlike the aerosol optical depth
35 cannot be constrained from space observations.

36
37 Dust particles are produced by disintegration of aggregates following creeping and saltation of larger soil
38 particles over desert and other arid surfaces (Kok, 2011; Zhao et al., 2006). The magnitude of dust emissions
39 to the atmosphere depends on the surface wind speed and many soil-related factors such as its texture,
40 moisture and vegetation cover. Since AR4, new dust emission schemes have been implemented in large-
41 scale models and compared against surface measurements and observation by remote sensing (Cheng et al.,
42 2008; Darmenova et al., 2009; Huneus et al., 2011b). The range of estimates for the global dust emission
43 span a factor of about 5 (Huneus et al., 2011b).

44
45 Dimethylsulphide (DMS) emitted from the oceans is a major natural contributor to the atmospheric sulphate
46 aerosol burden. Emissions of DMS to the atmosphere depend on its surface water concentration and its sea-
47 to-air transfer velocity. Since AR4, progress has been made in understanding how DMS surface water
48 concentrations are affected by solar radiation and atmospheric CO₂ concentration (Derevianko et al., 2009;
49 Kim et al., 2010). Prognostic models for predicting the spatial and temporal variation of the DMS surface
50 water concentration have been developed further and inter-compared for the first time (Le Clainche et al.,
51 2010). New DMS air-to-sea transfer models have been developed and evaluated (Huebert et al., 2010;
52 Vlahos and Monahan, 2009), and an updated climatology for surface ocean DMS concentrations and
53 emission fluxes has been developed (Lana et al., 2011). The range of the current estimates of global DMS
54 emissions span a factor of about 2. The majority of DMS emitted into the atmosphere is transformed to
55 particulate sulphate via gas-phase chemistry and cloud processing (Karl et al., 2007). Sulphate produced
56 from DMS has a major influence on marine CCN concentrations, but the relation between the DMS

emissions and CCN concentrations appears to be more complicated than previously thought (Korhonen et al., 2008; Thomas et al., 2010; Woodhouse et al., 2010).

Biogenic volatile organic compounds (BVOC) are globally the most important precursors for SOA. Emissions of BVOC to the atmosphere depend on the amount and type of vegetation, temperature, radiation and several environmental factors such as the ambient CO₂ concentration (Grote and Niinemets, 2008; Pacifico et al., 2009). While speciated BVOC emission inventories have been derived for some continental regions, global emission inventories or schemes are available only for isoprene and some monoterpenes (Guenther et al., 2006; Muller et al., 2008). The total global BVOC emissions have large uncertainties, despite the apparent convergence in different model-based estimates (Arneth et al., 2008). In the atmosphere, BVOCs are transformed to SOA with variable yields via gas-phase chemistry followed by gas-particle partitioning, and via other ageing mechanisms. The global SOA production resulting from BVOCs has been constrained substantially since AR4, the recent estimates being mostly within a factor 3 (Farina et al., 2010; Hallquist et al., 2009; Heald et al., 2010; Spracklen et al., 2011).

Formation of new particles with 1–2 nm diameters by nucleation and their initial growth to detectable sizes is called new particle formation (NPF). Atmospheric NPF has been observed to take place in a large number of different environments (Kulmala and Kerminen 2008; Manninen et al., 2010; O'Dowd et al., 2010) and substantial progress in our understanding has been made since AR4. Multiple lines of evidence indicate that sulfuric acid plays a central role in atmospheric NPF (Kerminen et al., 2010; Sipila et al., 2010), with observed particle formation rates scaling mostly to the power 1 to 2 of the gaseous sulphuric acid concentration (Kuang et al., 2008; Paasonen et al., 2010). Other compounds capable of influencing NPF include low-volatile organic vapors (Metzger et al., 2010; Paasonen et al., 2010; Wang et al., 2010b), ammonia and amines (Benson et al., 2011; Berndt et al., 2010; Kirkby et al., 2011; Kurten et al., 2008; Smith et al., 2010). Ion-induced nucleation is very likely to contribute to NPF throughout the atmosphere (Kirkby et al., 2011; Yu 2010), but NPF seems to be dominated by neutral nucleation pathways in continental boundary layers (Hirsikko et al., 2011; Kazil et al., 2010). NPF is not thought to affect directly the mass burden or direct RF of atmospheric aerosols. Model studies suggest, however, that NPF is a dominant source of the particle number concentration in the global atmosphere (Spracklen et al., 2006; Spracklen et al., 2010; Yu et al., 2010a) and a potentially significant contributor to global CCN (Kazil et al., 2010; Merikanto et al., 2009; Pierce and Adams 2009b). The growth of newly-formed particles to CCN sizes depends crucially on organic vapors (Riipinen et al., 2011), and is therefore strongly tied with atmospheric SOA formation.

Aerosol sinks are generally better understood than aerosol sources, and there has been correspondingly less progress in understanding and modeling since AR4. Improved dry deposition models, which depend the particle size as well as the roughness properties of the surface, have been parameterized and are increasingly being used in global aerosol models (Feng 2008; Kerkweg et al., 2006; Petroff and Zhang 2010). For the largest particles in the coarse mode, it is important to consider sedimentation throughout the atmosphere and its role in dry deposition at the surface. Aerosol wet deposition (in-cloud and below-cloud) is affected by the size and chemical composition of particles. For insoluble primary aerosol species like BC and dust, wet deposition depends also strongly on their degree of mixing with soluble species. Wet deposition of aerosols remain a key source of uncertainty in aerosol models which affects the vertical distribution and long-range transport of aerosols (Lee et al., 2011; Vignati et al., 2010). In particular, many models treat convective transport and scavenging sequentially rather than as a coupled process.

7.3.3 *Progresses and Gaps in Understanding Climate Relevant Aerosol Properties*

Since AR4, understanding some of the key aerosols properties has been greatly improved by laboratory and field experiments using advanced measurement technologies and theoretical studies. These experimental studies have stimulated improvement in the model representations of the aerosol physical, chemical and optical properties, and their role as cloud and ice nuclei (Figure 7.7; Table 7.2). Some of the field experiments have also been used to evaluate aerosol modules used in regional and global scale models. We focus our assessment on some of the key issues where there has been progress since AR4, focusing on BC, organic aerosols, aerosol size distribution and mixing state.

[INSERT TABLE 7.2 HERE]

Table 7.2: Key aerosol properties of the main aerosol species in the troposphere. Brown carbon is a particular type of OA but is treated here as an additional component because it is light absorbing. The estimate of aerosol burdens and lifetimes in the troposphere are based on the AeroCom models.

7.3.3.1 Chemical Composition

The gas-aerosol partitioning of semi-volatile inorganic species (H_2SO_4 , HNO_3 , HCl , and NH_3) is governed by gas-particle mass transfer kinetics, equilibrium thermodynamics, and heterogeneous chemical reactions. These processes are determined by meteorological conditions, such as temperature and relative humidity, and chemical conditions, such as gas-phase concentrations, aerosol size distribution and phase, and pH of particles. Aerosol models are increasingly calculating the gas-aerosol partitioning by assuming thermodynamic equilibrium (e.g., Nenes et al., 1998; Wexler and Clegg, 2002) or by solving gas-particle mass transfer dynamically (e.g., Zaveri et al., 2008; Zhang et al., 2004). Although a number of climate and global models assume thermodynamic equilibrium for semi-volatile inorganic species (or do not include semi-volatile inorganic species), the equilibrium assumption is not always valid even under typical atmospheric conditions.

BC is a distinct type of carbonaceous material, formed only near flames during combustion of carbon-based fuels. It strongly absorbs visible light and it is refractory. These physical properties allow at least in principle for a strict definition of BC. Direct measurement of individual BC-containing particles is possible with a single-particle soot photometer (SP2) based on laser-induced incandescence (Gao et al., 2007; Moteki and Kondo, 2010; Moteki et al., 2009; Schwarz et al., 2008; Schwarz et al., 2006). This technique has enabled accurate measurements of the size of BC cores (mean number diameters of 50–80 nm for fossil-fuel sources and 120 nm for biomass burning). However spatial and temporal coverage of the measurements are still limited (Schwarz et al., 2010).

Condensation of gas-phase species on BC and coagulation with other particles alter the mixing state of BC (e.g., Adachi et al., 2010; Li et al., 2003; Pósfai et al., 2003). In polluted urban air, BC becomes internally mixed on a timescale of ~ 12 h (McMeeking et al., 2010; Moteki et al., 2007). The resulting BC-containing particles become more hydrophilic which can lead to a reduced lifetime and atmospheric loading (Stier et al., 2006b). In addition, internal mixing can enhance the BC mass absorption efficiency by up to a factor of 2 (Bond and Bergstrom, 2006; Cross et al., 2010), with typical values of about $8\text{--}20 \text{ m}^2 \text{ g}^{-1}$ at a wavelength of 532 nm.

Long-range transport of BC has been observed in some conditions, e.g., from biomass burning in Russia to the Arctic in spring (Kondo et al., 2011; Warneke et al., 2009; Warneke et al., 2010), or in northeastern China in spring when humidity was relatively low (Oshima et al., 2011). However it is thought that rapid coating of BC by soluble material reduce the effectiveness of the vertical transport by convection.

Formation processes of OA remain highly uncertain, which is a major weakness in the present understanding of atmospheric aerosol (Hallquist et al., 2009; Kanakidou et al., 2005). Measurements by Aerosol Mass Spectrometers have provided some insights into sources of OA (Lanz et al., 2007; Ulbrich et al., 2009; Zhang et al., 2005b). Measurements at continental midlatitudes including urban and rural/remote air suggest that majority of SOA is likely to be oxygenated OA (Zhang et al., 2005a; Zhang et al., 2007). Measurements within and downstream of urban air indicate that under most circumstances SOA substantially contributes to the total OA mass (de Gouw et al., 2005; Volkamer et al., 2006; Zhang et al., 2007).

The contribution of secondary organic aerosol (SOA) to the total OA is larger than previously thought, but the split between primary organic aerosols (POA) and SOA has remained somewhat ambiguous (Jimenez et al., 2009; Robinson et al., 2007). POA had been assumed to be completely non-volatile. However, it has been found that a certain amount of POA may evaporate, followed by gas-phase photochemical oxidation of the vapors, to produce even less volatile SOA (Robinson et al., 2007), as shown in Figure 7.7.

Some of OA is light absorbing (brown carbon; BrC), which is produced mainly by lower temperature combustion, such as biomass burning (Andreae and Gelencser 2006). The absorption properties of BrC can be attributed to water soluble organic compounds and humic-like substances (Graber and Rudich 2006; Kirchstetter et al., 2004), although they are poorly quantified (Alexander et al., 2008; Flowers et al., 2010).

1
2 There is a large range in the complexity with which OA are represented in global aerosol models. Some
3 complex, yet still parameterized, chemical schemes have been developed recently which account for
4 multigenerational oxidation (Donahue et al., 2011; Jimenez et al., 2009; Robinson et al., 2007). Some
5 regional and global models use semi-empirical schemes, where semi- or non-volatile organic compounds
6 (SVOC) are produced from parent VOCs by oxidation processes and partitioned between the aerosol and gas
7 phases (Fan et al., 2005; Heald et al., 2005; Russell and Allen, 2005; Tsigaridis and Kanakidou, 2003). The
8 representation of secondary organic aerosols in many of the models used in CMIP5 is either non-existent or
9 very crude in that the source terms are prescribed and/or the models ignore the complex chemical and aging
10 processes.

11 7.3.3.2 *Mixing State*

12
13
14 There are multiple observations that show aerosols to be internally mixed (that is with multiple materials in
15 individual particles) relatively soon after emission. In general, organic and black carbon, in biomass burning
16 aerosols were found frequently internally mixed with ammonium, nitrate, and sulphate (Deboudt et al., 2010;
17 Pratt and Prather 2010). Studies over urban locations revealed that as much as 90% of the particles are
18 internally mixed with secondary inorganic species (Bi et al., 2011). Likewise there is evidence of internal
19 mixing between dust and biomass burning aerosols when these aerosol types age together (Hand et al.,
20 2010). Studies have shown that state of mixing can alter particle hygroscopicity and hence their ability to act
21 as CCN (Wex et al., 2010).

22
23 A common form of mixing is coating of soluble material (secondary aerosol) over a primary aerosol such as
24 a BC or dust core. The solid to liquid transition of such mixed aerosols as the RH increases influences their
25 light scattering properties through changes in particle shape, size, and refractive index (Freney et al., 2010).
26 Another form of coating involves fine BC contaminating large dust particles. The optical properties of an
27 internal mixture are known to be significantly different from those of an external mixture (Garcia et al.,
28 2011; Jacobson, 2001; Shiraiwa et al., 2008) with an increase in the mass absorption efficiency.

29
30 Global aerosol models are increasingly treating aerosols as an internal mixture, which enables a consistent
31 treatment of aerosol hygroscopicity, scavenging and optical properties (i.e., Stier et al., 2006a). Commonly-
32 used modal approaches do not allow to represent discrete variations of mixing state across an aerosol mode,
33 though.

34 7.3.3.3 *Aerosol Size Distribution*

35
36
37 Aerosol size distribution is one of the key characteristics required for estimating the spectral optical and
38 CCN properties of aerosols. Though ground-based measurements of aerosol size distribution are being
39 carried out as part of various field campaigns (Haywood et al., 2011), such information is still sparse and
40 there have been only few attempts to build a climatology of aerosol size distribution (e.g., Heintzenberg et
41 al., 2000). There have been improvements in the algorithms to derive column-averaged volume size
42 distribution from sun photometer measurements (Dubovik et al., 2006). Validation studies show agreement
43 for the derived size distribution against in situ (surface as well as aircraft-based) measurements
44 (Gerasopoulos et al., 2007; Haywood et al., 2011; Radhi et al., 2010; Smirnov et al., 2011), but these
45 inversion products have not been comprehensively validated. There has also been some progress in
46 measuring the aerosol size distribution down to a few nanometre size (e.g., Feldpausch et al., 2006) which
47 revealed large Aitken mode concentrations in the tropical upper troposphere.

48
49 Satellite sensors such as MODIS and POLDER also provides the Angström coefficient and AOD fine mode
50 fraction (i.e., contributed by those aerosols with diameter less than about 1 μm). Even though aerosol fine
51 mode fraction has been validated over ocean (Remer et al., 2005), accurate determination of fine mode
52 fraction over land still remains a challenge (Jethva et al., 2009; Levy et al., 2007).

53
54 Measurements have been used to evaluate the ability of the new generation of global aerosol models to
55 simulate aerosol number concentration and the aerosol size distribution (Bauer et al., 2008; Liu et al., 2005b;
56 Spracklen et al., 2005a, 2005b; Spracklen et al., 2010; Stier et al., 2005; Wang et al., 2009; Yu and Luo,
57 2009; Zhang et al., 2010b). Such evaluations place powerful constraints on the representation of new particle

1 formation in the models, which is a major source of uncertainty in estimates of the aerosol indirect effects
2 (see Section 7.4). Simulated aerosol number concentrations at the surface are usually found to be too low
3 unless empirical particle nucleation schemes are used. Models are also hampered by the lack of size-resolved
4 inventories of aerosol emissions.

6 7.3.3.4 *CCN Properties of Aerosols*

8 A proportion of aerosol acts as CCN. The ability of an aerosol particle to take up water and subsequently
9 activate is determined by its size and composition. Common CCN in the atmosphere are composed of sea
10 salt, sulphates and sulphuric acid, nitrate and some organics (Table 7.2). The size of the CCN was found to
11 be more important than their chemical composition at one continental location as larger particles are more
12 readily activated than smaller particles because they require a lower critical supersaturation (Dusek et al.,
13 2006). However, the chemical composition influences the aerosol size distribution and may be important in
14 other locations. The CCN activity of aerosols can be characterised by a hygroscopicity parameter κ that can
15 be derived from measurements (Petters and Kreidenweis, 2007).

16
17 CCN activity of inorganic aerosols is relatively well understood. Lately most attention has been paid to the
18 CCN activity of mixed organic/inorganic aerosols (e.g., King et al., 2010; Prisle et al., 2010). Uncertainties
19 in our current understanding of CCN properties are associated with SOA (Good et al., 2010), mainly because
20 OA are still poorly characterized (Jimenez et al., 2009). For SOA it is not clear how important surface
21 tension effects and bulk to surface partitioning are and if the water activity coefficient changes significantly
22 as a function of the solute concentration (Good et al., 2010; Prisle et al., 2008). Thus, when the aerosol is
23 dominated by organics, discrepancies between values of κ obtained directly from both CCN activity
24 measurements and sub-saturated particle water uptake measurements have been observed in some instances
25 (e.g., Irwin et al., 2010; Prenni et al., 2007; Roberts et al., 2010), whereas in other studies closure could be
26 obtained (e.g., Duplissy et al., 2008; Rose et al., 2011).

27
28 Pringle et al. (2010) used surface and aircraft measurements to evaluate the values of the hygroscopicity
29 parameter simulated by a global aerosol model, and found generally good agreement. Spracklen et al. (2008)
30 used surface and aircraft measurements to evaluate simulated distributions of CCN concentration, and found
31 good agreement between simulated and observed concentrations.

33 7.3.3.5 *IN Properties of Aerosols*

34
35 Aerosols that act as IN are solid substances at atmospheric temperatures and supersaturations. Mineral dust
36 and primary biological particles are typically known as good IN. Conflicting evidence has been presented for
37 the ability of BC, organic and biomass burning particles to act as IN. Four heterogeneous ice nucleation
38 modes are distinguished in the literature. Immersion freezing refers to freezing that is initiated from within a
39 cloud droplet, condensation freezing refers to freezing during droplet formation, and contact freezing is
40 initiated when an IN collides with a supercooled cloud droplet. Deposition nucleation refers to the direct
41 deposition of vapour onto IN. Lidar observations revealed that liquid cloud droplets are present before ice
42 crystals form via heterogeneous freezing mechanisms (Ansmann et al., 2008; de Boer et al., 2011) indicating
43 that deposition nucleation does not seem to be important for mixed-phase clouds.

44
45 A compilation of onset temperatures and relative humidities for deposition/condensation nucleation versus
46 immersion freezing for bacteria, mineral dust and soot based on recent data from laboratory data is shown in
47 Figure 7.8. It should be noted that the reported nucleation onset points for the different materials differ with
48 respect to the fraction of ice nucleating particles (e.g., due to the detection thresholds of measurement
49 methods). Bacteria initiate immersion freezing at the highest temperatures, followed by mineral dust and
50 then soot. It is obvious that for dust and bacteria, heterogeneous ice nucleation occurs at considerably higher
51 temperatures than homogeneous freezing. Remote sensing observations confirm that ice clouds in air
52 containing dust can be found at significant warmer temperatures than in dust-free conditions (Choi et al.,
53 2010; Sassen et al., 2003; Seifert et al., 2010). Laboratory results indicate that, in comparison with natural IN
54 such as mineral dust and biological particles, soot initiates ice at the coldest temperatures. Deposition
55 nucleation of ice on most types of soot particles is not important above -30°C and below water saturation
56 (Dymarska et al., 2006; Friedman et al., 2011). However, in-situ observations indicate an enrichment of BC

1 in atmospheric ice particle residuals in tropospheric mixed phase clouds (Cozic et al., 2008; Targino et al.,
2 2009; Twohy et al., 2010), thus there must be some mechanism for BC to enter ice clouds.

3
4 Lately, a variety of other substances such as crystalline ammonium sulphate at cirrus temperatures (e.g.,
5 Abbatt et al., 2006; Baustian et al., 2010; Wise et al., 2009), oxalic acid (Wagner et al., 2010, 2011; Zobrist
6 et al., 2006), marine diatoms (Knopf et al., 2011) as well as fulvic and humic acid (Wang and Knopf, 2011)
7 have been identified as possible IN. Biological particles in significant concentrations have also been
8 observed in residues from ice crystals (Pratt et al., 2009; Prenni et al., 2009).

9
10 IN can either be bare or mixed with other substances. As bare particles age in the atmosphere, they acquire
11 liquid surface coatings by condensing soluble species and water vapor or by scavenging soluble particles,
12 which may transform IN from deposition or contact nuclei into possible immersion nuclei. This
13 transformation or the chemical processing with ozone or other oxidizing substances may dampen the ice-
14 forming ability of some IN types (Chernoff and Bertram, 2010; Cziczo et al., 2009b; DeMott et al., 2009;
15 Eastwood et al., 2009; Mohler et al., 2005; Sullivan et al., 2010b; Wang and Knopf, 2011). IN have been
16 observed to be less efficient after chemical aging with nitric acid (Sullivan et al., 2010a) with ammonia gas
17 (Salam et al., 2007) and of BC particles with aqueous sulphuric acid for nucleation in cirrus clouds (DeMott
18 et al., 1999; Koehler et al., 2009).

19 [INSERT FIGURE 7.8 HERE]

20 **Figure 7.8:** The onset temperatures and relative humidities for deposition/condensation freezing and immersion
21 freezing for bioaerosols (Ahern et al., 2007; Diehl et al., 2001; Iannone et al., 2011; Kanji et al., 2011; Mohler et al.,
22 2008; Mortazavi et al., 2008; von Blohn et al., 2005; Yankofsky et al., 1981), mineral dusts (Archuleta et al., 2005;
23 Bundke et al., 2008; Connolly et al., 2009; Cziczo et al., 2009a; Field et al., 2006; Kanji and Abbatt 2006; Kanji et al.,
24 2011; Knopf and Koop 2006; Koehler et al., 2010; Kulkarni and Dobbie 2010; Lüönd et al., 2010; Mohler et al., 2006;
25 Murray et al., 2011; Niedermeier et al., 2010; Niemand et al., 2011; Roberts and Hallett 1968; Salam et al., 2006;
26 Schaller and Fukuta 1979; Welte et al., 2009; Zimmermann et al., 2008), organics (Baustian et al., 2010; Kanji et al.,
27 2008; Petters et al., 2009; Prenni et al., 2007; Shilling et al., 2006; Wagner et al., 2010, 2011; Wang and Knopf 2011;
28 Zobrist et al., 2007), solid ammonium sulphate (Abbatt et al., 2006; Baustian et al., 2010; Mangold et al., 2005; Shilling
29 et al., 2006; Wise et al., 2009; 2010) and BC (soot) (Crawford et al., 2011; DeMott 1990; DeMott et al., 1999; Diehl
30 and Mitra 1998; Dymarska et al., 2006; Fornea et al., 2009; Gorbunov et al., 2001; Kanji et al., 2011; Mohler et al.,
31 2005), from a compilation of experimental data of sub- and super-micron aerosol particles in the literature (for
32 references see supplementary material). The large range of observed ice nucleation onset conditions is due to different
33 experimental setups, particle sizes, activated fractions and chemical composition. Only those IN species for which at
34 least three papers exists are shown. The dashed line refers to the homogeneous freezing of solution droplets after (Koop
35 et al., 2000).
36

37 7.3.4 Aerosol Distributions

38 This section assesses our current understanding of aerosol distributions from in-situ and remote-sensing
39 measurements, and the current ability of global aerosol models to simulate such distributions. Aerosol
40 predictions in the CMIP5 models are assessed in Chapter 12. Since the AR4, new and improved
41 observational tools for global aerosol model evaluation have emerged.
42

43 The most commonly used in-situ measurements for global aerosol model evaluation are mass concentrations
44 and deposition fluxes. Long-term aerosol mass concentrations have been measured at the surface by global
45 and local networks, as well as individual efforts with improved coordination and quality assurance.
46 Climatologies of the main aerosol types can be constructed from such measurements (e.g., Jimenez et al.,
47 2009; Figure 7.9). They show a wide spatial variability in the dominant aerosol type and aerosol
48 concentrations worldwide. Mineral aerosol is the largest aerosol component in most areas with higher
49 concentrations in Urban S. Asia and China, accounting ~ 35% of PM₁₀. Aerosol fraction in rural U.S. and
50 South America are composed mainly (i.e., ~20%) of OC, and the OC fractions also rank second or third with
51 a mean of ~16% in other areas of the world. Sulfate normally accounts for ~10–30%, except for the areas in
52 rural Africa, high Asia, urban Oceania and South America with less than ~5%. The fractions of nitrate and
53 ammonium are only around 6% and 4%, in average respectively. In most areas, BC fractions are less than
54 5%, although this percentage may be larger in South America, urban Africa, urban Europe, South-East and
55 East Asia and urban Oceania. Sea-salt aerosols can be dominant at oceanic sites. Measurements of dry and
56 wet deposition on the surface have also been made (e.g., Hjellbrekke, 2001; McConnell et al., 2007) and
57
58

1 used to evaluate global aerosol models (e.g., Boucher and Pham, 2002; Easter et al., 2004; Flanner et al.,
2 2007; Huneus et al., 2011b).

3 4 **[INSERT FIGURE 7.9 HERE]**

5 **Figure 7.9:** Bar chart plots summarizing the annual, seasonal or monthly mean mass concentration ($\mu\text{g m}^{-3}$) of six
6 major types of aerosol particles in diameter smaller than 10 μm with at least an entire year data from various rural and
7 urban sites in nine continental areas of the world. These include: 1) rural U. S. (Chow et al., 1993; Liu et al., 2005a;
8 Malm and Schichtel 2004; Malm et al., 1994); urban U. S. (Chow et al., 1993; Ito et al., 2004; Kim et al., 2000; Liu et
9 al., 2005a; Malm and Schichtel 2004; Sawant et al., 2004); 2) South America (Artaxo et al., 1998; Artaxo et al., 2002;
10 Bourotte et al., 2007; Celis et al., 2004; Fuzzi et al., 2007; Gioda et al., 2011; Mariani and Mello 2007; Martin et al.,
11 2010; Morales et al., 1998; Souza et al., 2010); 3) rural Europe (Gullu et al., 2000; Hueglin et al., 2005; Kocak et al.,
12 2007; Putaud et al., 2004; Puxbaum et al., 2004; Querol et al., 2001; Querol et al., 2009; Querol et al., 2004; Rodriguez
13 et al., 2002; Rodriguez et al., 2004; Salvador et al., 2007; Theodosi et al., 2010; Viana et al., 2008; Yin and Harrison
14 2008; Yttri 2007); urban Europe (Hueglin et al., 2005; Lenschow et al., 2001; Lodhi et al., 2009; Lonati et al., 2005;
15 Perez et al., 2008; Putaud et al., 2004; Querol et al., 2001; Querol et al., 2006; Querol et al., 2004; Querol et al., 2008;
16 Rodriguez et al., 2002; Rodriguez et al., 2004; Roosli et al., 2001; Viana et al., 2007; Viana et al., 2006; Yin and
17 Harrison 2008); 4) rural Africa (Maenhaut et al., 1996; Mkoma 2008; Mkoma et al., 2009a; Mkoma et al., 2009b;
18 Nyanganyura et al., 2007; Weinstein et al., 2010); urban Africa (Favez et al., 2008; Mkoma 2008; Mkoma et al.,
19 2009a); 5) high Asia, with altitude larger than 1680 m. (Carrico et al., 2003; Decesari et al., 2010; Ming et al., 2007a;
20 Qu et al., 2008; Ram et al., 2010; Rastogi and Sarin 2005; Rengarajan et al., 2007; Shresth et al., 2000; Zhang et al.,
21 2001; Zhang et al., 2008; Zhang et al., 2011a); 6) rural China (Hagler et al., 2006; Hu et al., 2002; Zhang et al., 2011a);
22 urban China (Cheng et al., 2000; Hagler et al., 2006; Oanh et al., 2006; Wang et al., 2003; Wang et al., 2005b; Wang et
23 al., 2006; Xiao and Liu 2004; Yao et al., 2002; Ye et al., 2003; Zhang et al., 2002; Zhang et al., 2011a; Zhang et al.,
24 2011b); 7) South-East and East Asia (Han et al., 2008; Khan et al., 2010; Kim et al., 2007; Lee and Kang 2001; Oanh et
25 al., 2006); 8) urban South Asia (Chakraborty and Gupta 2010; Khare and Baruah 2010; Kumar et al., 2007; Lodhi et al.,
26 2009; Raman et al., 2010; Rastogi and Sarin 2005; Safai et al., 2010; Stone et al., 2010); 9) urban Oceania (Chan et al.,
27 1997; Maenhaut et al., 2000; Radhi et al., 2010; Wang and Shooter 2001; Wang et al., 2005a).

28
29 Measurements of aerosol optical depth (AOD) are retrieved at the surface by AERONET (e.g., Holben et al.,
30 1998; Holben et al., 2001), other ground-based networks and an increasing number of satellite missions.
31 Retrievals from aerosol-dedicated instruments such as MODIS (Kleidman et al., 2011; Levy et al., 2010;
32 Remer et al., 2005), MISR (Kahn et al., 2005; Kahn et al., 2007) and POLDER/PARASOL (Tanré et al.,
33 2011). Other instruments such as AVHRR (Geogdzhayev et al., 2002; Jeong and Li 2005), TOMS (Torres et
34 al., 1998; Torres et al., 2002) and ATSR/AATSR have longer measurements records. Composite aerosol
35 datasets have also been developed (see Chapter 2). While each aerosol retrieval shows some skill against
36 AERONET measurements, there are still large differences in regional and seasonal patterns because of
37 differences in sampling, cloud screening and treatment of the surface reflectivity (Kokhanovsky et al., 2010).

38
39 Field experiments involving research aircraft such as TRACE-P (Jacob et al., 2003), INTEX-A (Singh et al.,
40 2006), ARCTAS (Jacob et al., 2010), ARCPAC (Warneke et al., 2010) and HIPPO1 (Schwarz et al., 2010;
41 Schwarz et al., 2006), commercial aircraft (Brenninkmeijer et al., 2007) and spaceborne lidars (Omar et al.,
42 2009; Winker et al., 2009) can provide measurements of the aerosol vertical profile. For example, Koch et al.
43 (2009b) and Schwarz et al. (2010) used BC measurements by an SP2 instrument on aircraft to evaluate
44 AeroCom model simulations of the vertical distribution of BC aerosol in many regions, and found that most
45 models simulate too much BC in the upper troposphere (Schwarz et al., 2010, see Figure 7.10). Koch et al.
46 (2009b) also used AERONET retrievals of aerosol absorption optical depth (AAOD) to show that most
47 AeroCom models underestimate AAOD in many regions. Yu et al. (2010b) and Koffi et al. (2011) found that
48 global aerosol models tend to have a positive bias on the aerosol extinction scale height in some (but not all)
49 regions resulting in an overestimate of aerosol concentrations above 6 km.

50 51 **[INSERT FIGURE 7.10 HERE]**

52 **Figure 7.10:** Comparison of BC profiles as measured during the ARCTAS, HIPPO1 and FORCE-A campaigns and
53 simulated by a range of global aerosol models. [PLACEHOLDER FOR SECOND ORDER DRAFT: will be updated
54 from AeroCom and CMIP5 models]

55
56 Overall aerosol measurements have been widely used in the evaluation of aerosol models both within the
57 AeroCom activity (e.g., Huneus et al., 2011b; Kinne et al., 2006; Koch et al., 2009b) and elsewhere (e.g.,
58 Mann et al., 2010; Myhre et al., 2007; Wang et al., 2009). This has contributed to continuous model
59 improvement since AR4 but models are still imperfect [PLACEHOLDER FOR SECOND ORDER DRAFT:

1 AeroCom reference]. In a few studies the measurements are also used directly to improve aerosol
2 distributions through data assimilation (e.g., Benedetti et al., 2009; Huneus et al., 2011a). The lack of an
3 observational constraint on the pre-industrial aerosol distribution is a continuous source of uncertainty when
4 estimating aerosol radiative forcing.

5 6 **7.3.5 Aerosol Radiative Effects**

7 8 *7.3.5.1 Direct Radiative Effect*

9
10 Direct radiative effect (DRE) is the change in radiative flux caused by the combined direct effect of all
11 anthropogenic and natural aerosols. The physics behind the DRE is robust and the DRE is close to being an
12 observable quantity, yet our knowledge of aerosol and environmental characteristics needed to quantify the
13 DRE at the global scale remains incomplete (Anderson et al., 2005; Jaegle et al., 2011; Satheesh and
14 Moorthy 2005). The DRE of aerosols depends on the distribution of their optical properties and the
15 scattering and absorbing properties of the environment (i.e., surface, atmospheric molecules and clouds) as
16 illustrated on Figure 7.7. More specifically the DRE requires the knowledge of the spectral variations in
17 aerosol extinction coefficient, single scattering albedo and phase function, which can in principle be
18 estimated from the aerosol size distribution, shape, chemical composition and the state of mixing. In the
19 solar spectrum, the DRE is typically negative at the top-of-atmosphere but gets weaker and can become
20 positive with increasing aerosol absorption, decreasing upscatter fraction, or increasing albedo of the
21 underlying surface. DRE is largest in cloud-free conditions, but can be significant in the presence of a thin
22 cloud layer. Top-of-atmosphere DRE can become positive when the aerosols are located above clouds (e.g.,
23 Chand et al., 2009). The DRE at the surface is almost always negative and its magnitude can be much larger
24 than the DRE at top-of-atmosphere when there is aerosol absorption (Li et al., 2010). In the longwave
25 spectrum, the DRE is generally positive at the top-of-atmosphere but is only significant for desert dust and
26 sea-salt (Reddy et al., 2005).

27
28 There have been many measurement-based estimates of the DRE (Bauer et al., 2011; Bergamo et al., 2008;
29 Di Biagio et al., 2010; Yu et al., 2006) although some studies involve some degree of modelling. There is
30 generally a good agreement between observed and calculated shortwave radiative fluxes when aerosol
31 properties are known (e.g., Osborne et al., 2011). Global observational estimates of the DRE rely on satellite
32 remote sensing of aerosol properties and/or measurements of the Earth's radiative budget (Chen et al., 2011;
33 Haywood et al., 2011). The shortwave clear-sky top-of-atmosphere DRE over the ocean is estimated to be in
34 a range from -4 to -6 W m^{-2} , with sea-salt being the main contributor (Bellouin et al., 2005; Loeb and
35 Manalo-Smith 2005; Myhre et al., 2007; Yu et al., 2006). Uncertainties are larger over land because satellite
36 retrievals are more difficult and the surface is less well characterised (Chen et al., 2009; Jethva et al., 2009)
37 despite recent progress in inversion algorithms (e.g., Dubovik et al., 2011). Attempts to estimate the DRE in
38 cloudy-sky remain elusive (e.g., Peters et al., 2011) although remote sensing of aerosols over clouds is now
39 possible from passive (Waquet et al., 2009) and active (Omar et al., 2009) methods. Notable areas of positive
40 DRE include the Arctic, where absorbing fossil-fuel aerosols overlie ice surfaces (Stone et al., 2008), and off
41 the shore of Namibia, where absorbing biomass-burning aerosols seasonally overlie stratocumulus clouds.
42 While AOD and aerosols size are relatively well constrained, uncertainties in the aerosol single-scattering
43 albedo (Loeb and Su, 2010) and vertical profile (e.g., Zarzycki and Bond, 2010) contribute significantly to
44 the overall uncertainties in DRE, especially for all-sky estimates.

45 46 *7.3.5.2 Aerosol Semi-Direct Effect and its Impact on Precipitation*

47
48 BC, dust and other absorbing aerosols perturb the temperature structure of the atmosphere through radiative
49 heating, modify the surface fluxes, and thus can influence cloud cover, precipitation and atmospheric
50 dynamics. This effect, known as the semi-direct effect, can be considered as a rapid response associated to
51 the direct effect and as such it can be accounted for through the concept of AF introduced in Chapters 1 and
52 8. Denman et al. (2007) assessed the semi-direct aerosol forcing to be small and of indeterminate sign and
53 attached a very low understanding to it. This very low understanding resulted from the differences between
54 cloud resolving and global model studies.

55
56 Since AR4 the observational evidence for the semi-direct effect has strengthened. There are additional
57 observations of variations in cloud cover correlating with variations in the amount of absorbing aerosols

1 (e.g., Brioude et al., 2009; Wilcox, 2010) for which the semi-direct effect has been offered as an explanation.
2 Such a causal effect between aerosol atmospheric heating (and surface cooling) and cloudiness has been
3 confirmed by modelling studies on eddy-resolving, regional and global scale, which have revealed a more
4 complicated picture than initially anticipated (e.g., Hill and Dobbie, 2008; Sakaeda et al., 2011; Zhuang et
5 al., 2010).

6
7 Absorbing aerosol modifies atmospheric stability; the effect of this on cloud cover depends on the height of
8 the aerosol relative to the clouds (Allen and Sherwood, 2010; Yoshimori and Broccoli, 2008). Both
9 scattering and absorbing aerosols are very effective at reducing the downwelling solar radiation at the
10 surface such that their changes contribute to solar dimming and brightening (see also Chapter 2). Balance in
11 the net surface radiation therefore requires that either the upwelling surface sensible or latent heat fluxes
12 decrease. Over land, where surface heating is a primary driver for convective clouds, this will affect cloud
13 fraction and depth. Together the mechanisms of stabilisation and reduction in surface fluxes provide a means
14 for aerosols to significantly modify the cloud fraction of surface-forced continental clouds (Feingold et al.,
15 2005; Sakaeda et al., 2011). The microphysical properties of the cloud can modify entrainment rates and
16 have also been shown to affect the semi direct effect when BC occurs within the cloud layer (Hill and
17 Dobbie, 2008).

18
19 Cloud cover is expected to decrease if absorbing aerosol is embedded in the cloud layer. This has been
20 observed (Koren et al., 2004) and simulated (e.g., Feingold et al., 2005) for clouds over the Amazon forest in
21 the presence of smoke aerosols. In the stratocumulus regime, absorbing aerosol above cloud-top strengthens
22 the temperature inversion, reduces entrainment and tends to enhance cloudiness. Satellite observations
23 (Wilcox, 2010) and modelling (Johnson et al., 2004) of marine stratocumulus show a thickening of the cloud
24 layer beneath layers of absorbing smoke aerosol, which gives a local negative semi-direct effect.

25
26 GCMs lack the ability of large eddy simulation models to represent smaller scale cloud processes and this
27 gives low confidence in their ability to simulate the semi-direct effect (Johnson, 2005). Conversely, the
28 GCM response allows for large scale circulation changes that can have significant additional effects which
29 are not captured in large eddy models. Importantly, GCM results have shown that semi-direct effects are not
30 confined to absorbing aerosol, CO₂ and potentially all mechanisms have an associated tropospheric
31 adjustment (Section 7.2.4.3.6). Further, the semi-direct effect is not only associated with cloud changes that
32 are co-located with the forcing, it also leads to circulation changes that effect clouds and land surface
33 properties remotely.

34
35 There is a clear link between changes in atmospheric heating and global mean precipitation changes
36 (Andrews et al., 2010; Lambert and Allen, 2009; Liepert and Previdi, 2009; Stephens and Ellis, 2008;
37 Takahashi, 2009; Wild and Liepert, 2010). Andrews et al. (2010) showed that absorbing aerosols lead to an
38 initial reduction in global precipitation which is not completely offset by the slow global precipitation
39 change that responds linearly to the global mean surface temperature change.

40 41 7.3.5.3 *Estimates of Aerosol Radiative and Adjusted Forcings*

42
43 Building on our understanding of aerosol concentration distributions (Section 7.3.4) and their radiative
44 effects (Sections 7.3.5.1 and 7.3.5.2), the section combines the direct RF with the adjustment effect to
45 produce an AF. The subsection additionally assesses the forcings from absorbing aerosol (BC and dust) on
46 both snow and ice.

47
48 In Forster et al. (2007) these adjustment effects were not included in the forcing term but were evaluated as
49 part of the response. Instead, adjustment was accounted for by applying an efficacy term that modified the
50 climate sensitivity. For consistency with Chapter 8, all quoted ranges represent a 2- σ uncertainty (i.e., 5% to
51 95% probability and we evaluate radiative forcings between 1750 and ~2010. Note that for several aerosol
52 species (such as biomass burning) this does not quite equate to the anthropogenic effect as emissions started
53 to be influenced by humans before the industrial revolution. Many models estimate the aerosol radiative
54 forcings between 1850 and present-day and conversion to a radiative forcing between 1750 and present-day
55 contributes to increase the uncertainty (Bellouin et al., 2008).

1 The estimate of the total aerosol direct radiative forcing in Forster et al. (2007) combined estimates from 9
2 AeroCom models, 8 other model studies and 3 observationally-based estimates. The model direct radiative
3 forcing was estimated to be -0.4 W m^{-2} with a 0 to -0.8 W m^{-2} range. However, the observationally-based
4 estimates gave a more negative radiative forcing, so overall a best estimate for the aerosol direct forcing was
5 given as $-0.5 \pm 0.4 \text{ W m}^{-2}$. Due to the extra evidence from observations for the total forcing there was more
6 confidence placed in the total aerosol direct forcing than in that from individual aerosol species.

7
8 Observationally-based estimates of the direct RF are not completely independent of global aerosol models
9 (Loeb and Su, 2010; Myhre, 2009). They employ satellite data of aerosol optical depth in combination with
10 either aerosol optical properties from AERONET (Bellouin et al., 2008) or observationally-derived aerosol
11 radiative efficiency (Quaas et al., 2008). In the observationally-based method, satellite retrieval constraints
12 over land necessitate the use of information from global aerosol models in order to derive the change in
13 AOD that is due to anthropogenic activity. The aerosol optical properties (single scattering albedo and
14 asymmetry factor) have been held constant between preindustrial and present time, and radiative forcing
15 calculations have been performed with radiative transfer schemes (Bellouin et al., 2008), or the aerosol
16 radiative efficiency has been assumed to be constant over the industrial era (Quaas et al., 2008).

17
18 Since AR4 further observationally-based estimates of the total direct radiative forcing have been made.
19 These employ both improved observations and also a more robust methodology for combining with models
20 to infer radiative forcing. Zhao et al. (2008) derived a clear-sky anthropogenic aerosol direct forcing over
21 ocean from a combination of CERES/MODIS data and the GOCART aerosol model finding radiative effects
22 that would broadly agree with equivalent estimates from Bellouin et al., (2008). Two studies (Bellouin et al.,
23 2008; Myhre, 2009) identified a number of causes for the larger magnitude in the observationally-based
24 radiative forcing estimate compared with modelling-based estimates within AR4. Compared to Bellouin et
25 al., (2005), Bellouin et al., (2008) refined their calculation of the direct radiative forcing using updated
26 satellite retrievals, finding an all sky radiative forcing of -0.65 W m^{-2} . They were able to attribute a more
27 negative observationally-based forcing to i) a positive cloudy-sky direct radiative forcing in the model, ii) the
28 exclusion of high surface albedo regions, and iii) the fact that the model forcing was compared to 1750 rather
29 than present day natural aerosol for the observed estimate. Myhre (2009) extended this analysis and used
30 model results to adjust their observed estimate to make it representative of a globally averaged all-sky
31 present – preindustrial radiative forcing. Making these changes reduced the magnitude of the
32 observationally-based radiative forcing estimate from -0.65 W m^{-2} to $-0.3 \pm 0.2 \text{ W m}^{-2}$. An additional
33 comparison of the Oslo model with various observations suggested that the estimate of total direct RF was
34 rather robust, giving increased confidence to their estimate.

35
36 Loeb and Su (2010) have challenged the uncertainty ranges presented in Forster et al. (2007) and Myhre
37 (2009). They use an estimate of present-day AERONET uncertainties in aerosol optical depth, single-
38 scattering albedo, asymmetry parameter, aerosol scale height, and anthropogenic fraction. The aerosol
39 optical depth was perturbed by ± 0.01 , the asymmetry parameter was perturbed by ± 0.02 , and the single-
40 scattering albedo was perturbed by ± 0.06 over ocean and ± 0.03 over land. The scale height was perturbed by
41 0.8 km and the anthropogenic fraction by 10%. Their analysis does not take into account additional
42 uncertainties in the estimate of direct radiative forcing such as preindustrial emissions. Nevertheless, they
43 find that uncertainties in aerosol optical depth alone can lead to errors of $\pm 0.2 \text{ W m}^{-2}$. The single-scattering
44 albedo error was found to be much more significant, contributing an error of greater than $\pm 0.5 \text{ W m}^{-2}$. Our
45 own assessment of uncertainty (see below) falls between those from past studies, indicating that the Loeb
46 and Su (2010) approach is valid but likely over-estimated the single scattering albedo uncertainty.

47
48 Lohmann et al. (2010) found direct RFs in five GCMs ranged from -0.1 to -0.4 W m^{-2} . A second phase of
49 AeroCom model results is being compiled, based on updated emissions and model codes. Simulations of the
50 secondary components nitrate and SOA are now included in the analysis. Estimates of the radiative forcing
51 for the total anthropogenic direct aerosol effect range from -0.06 to -0.49 W m^{-2} . Figure 7.11 shows the
52 zonal mean total aerosol RF for all participating AeroCom models. Most of the models have a maximum
53 negative radiative forcing around $20\text{--}50^\circ\text{N}$, in the region with highest aerosol concentrations. Several models
54 show a positive radiative forcing at high latitudes due to the higher surface albedo there. We combine our
55 uncertainty analysis with the best estimate of -0.3 W m^{-2} from the satellite derived estimates, reinforced by
56 the RF from the Lohmann et al. (2010) and the new AeroCom modelling studies, to estimate an aerosol
57 direct RF of $-0.3 \pm 0.3 \text{ W m}^{-2}$.

1
2 **[INSERT FIGURE 7.11 HERE]**

3 **Figure 7.11:** Zonal mean total aerosol direct radiative forcing from the different AeroCom models. No adjustment for
4 missing species has been applied.

5
6 The semi-direct effect in GCMs has been estimated from both fixed sea surface temperature experiments and
7 regression methods (e.g., Allen and Sherwood, 2010; Andrews and Forster, 2008; Lohmann et al., 2010).
8 Most GCM studies indicate regional variations in the cloud response but generally increased cloud cover
9 over oceans, especially in low and mid level clouds (Allen and Sherwood, 2010; Sakaeda et al., 2011). Over
10 land the response is more dependent on the type of forcing (Allen and Sherwood, 2010; Koch and Del Genio,
11 2010; Sakaeda et al., 2011). Overall the result is thought to be a small and possibly net negative semi-direct
12 effect feedback from the cloud response to absorbing aerosols (Koch and Del Genio, 2010). Five GCMs
13 were analysed for RF and AF in Lohmann et al. (2010). One GCM (CSIRO) had a large difference between
14 its RF and AF, giving a significant and negative semi-direct effect for the direct aerosol forcing of around –
15 0.3 W m^{-2} , traced to a longwave cloud adjustment. The other four GCMs analysed exhibited both positive
16 and negative semi-direct effects but none were significant. Based on current understanding, there is high
17 confidence at the local scale that in-situ heating by absorbing aerosol can cause cloud to both increase and
18 decrease, depending on specific conditions. However, there is low confidence in a globally-significant effect
19 as GCMs differ in their responses and are not able to adequately represent some of the important cloud
20 processes. Thus the semi-direct effect can be significant regionally and can be of either sign. Globally the
21 semi-direct effect is likely to contribute an AF that is small (smaller than 0.1 W m^{-2}) and not significantly
22 different than zero. In conclusion, adding the semi-direct effect to the RF increases the uncertainty range, but
23 does not alter the best estimate, which gives an AF of $-0.3 \pm 0.4 \text{ W m}^{-2}$.

24 25 7.3.5.4 *Aerosol Direct Radiative Forcing by Species*

26
27 AeroCom studies have calculated the direct radiative forcing using preindustrial and present-day simulations
28 with the same meteorology and no microphysical changes to isolate direct effects. For the RF of the
29 individual species (SO_4 , BC fossil-fuel, OC fossil-fuel, biomass burning or BB, SOA, NO_3) simulations are
30 performed as a difference between the control and a new simulation with only the aerosol component in
31 question set to emissions as in the preindustrial simulation (Figure 7.12).

32
33 For sulphate AeroCom models give a RF of -0.2 to -0.6 W m^{-2} 90% range with a -0.3 W m^{-2} median
34 estimate and we adopt this our our best estimate.

35
36 Ramanathan and Carmichael (2008) suggest a radiative effect of $+0.9 \text{ W m}^{-2}$ for the total (anthropogenic
37 plus natural) BC using an analysis of AERONET data. However, they likely overestimate the aerosol optical
38 depth as their results may be contaminated by dust. The current AeroCom models have components of BC
39 from fossil fuel and biomass burning. We take the estimate of RF from BC aerosol from fossil fuel from
40 AeroCom as $+0.0$ to $+0.4 \text{ W m}^{-2}$ 90% range with a $+0.2 \text{ W m}^{-2}$ median estimate. Zarzycki and Bond (2010)
41 assess uncertainty in the BC forcing-efficiency associated with the vertical profile of BC. They find that
42 uncertainty in the vertical profile contributes an uncertainty of around 25% to the forcing but also a positive
43 bias of around 15% in model-estimated radiative forcing, as models tend to have too much BC in the upper
44 troposphere compared to observations (see Section 7.3.4). When the AeroCom BC fossil fuel and BC from
45 biomass burning are combined and scaled by AERONET observations, we derive an anthropogenic BC
46 radiative forcing of $0.4 \pm 0.2 \text{ W m}^{-2}$, considering uncertainties associated with model diversity, AERONET-
47 model bias, AERONET representativeness and clear sky bias (e.g., AERONET does not observe over ocean
48 or on cloudy days), dust contamination of AERONET data, anthropogenic fraction, vertical distribution of
49 BC, underlying surface albedo, radiative transfer and covariance of aerosol with clouds.

50
51 For organic carbon aerosol from fossil fuel AeroCom models give a -0.0 to -0.1 W m^{-2} (90% range) with a $-$
52 0.05 W m^{-2} median estimate from fossil fuel emissions and we adopt this our our best estimate.

53
54 For biomass burning aerosol AeroCom models give a -0.15 to $+0.1 \text{ W m}^{-2}$ (90% range) with a -0.01 W m^{-2}
55 median estimate and we adopt this our our best estimate.

56

1 For SOA AeroCom models give a -0.03 to -0.07 W m^{-2} 90% range with a -0.04 W m^{-2} median estimate and
2 we adopt this our our best estimate.

3
4 We combine the AeroCom range with earlier estimates from Adams et al. (2001), Bauer et al. (2007) and
5 Myhre et al. (2009) give a RF estimate of -0.1 ± 0.08 W m^{-2} for nitrate aerosols.

6
7 Anthropogenic sources of mineral aerosols can result from changes in land use and water use or climate
8 change. Estimates of the anthropogenic radiative forcing of mineral aerosols is highly uncertain, because
9 natural and anthropogenic sources of mineral aerosols are often located close to each other (Mahowald et al.,
10 2009). Using a compilation of observations of dust records over the 20th century with model simulations,
11 Mahowald et al. (2010) deduce an 1750–2000 change in mineral aerosol direct radiative forcing including
12 both natural and anthropogenic changes of -0.14 ± 0.11 W m^{-2} . This is consistent within the AR4 estimate of
13 -0.1 ± 0.2 W m^{-2} (Forster et al., 2007) which we retain here. Note that some of this forcing could be due to
14 feedback processes (see Section 7.3.6).

15 [INSERT FIGURE 7.12 HERE]

16 **Figure 7.12:** Median, full range and 5%–95% range of AeroCom model direct radiative forcing by species and the total
17 direct forcing. The total direct forcing has been adjusted to take account of missing species in some models by adding
18 the median value of the species forcing from the remaining models.

19 7.3.5.5 *Absorbing Aerosol on Snow and Sea-Ice*

20
21 Forster et al. (2007) estimated the radiative forcing for surface albedo changes associated with BC on snow
22 to be 0.10 ± 0.10 W m^{-2} , with a low level of understanding. This estimate was largely based on calculations
23 with the GISS climate model (Hansen and Nazarenko, 2004) and a prognostic estimate by Jacobson (2004).
24 Since AR4, understanding, observations and modelling have all improved such that a more robust
25 assessment can be made. Further, additional effects of BC in snow and ice have been observed and
26 estimated. The role of non-BC constituents has also been investigated.

27
28 Global model studies have either scaled albedo changes based on model-derived BC deposition rates
29 (Hansen et al., 2005; Shindell and Faluvegi, 2009) or have prognostically determined the concentrations of
30 BC in snow (Flanner et al., 2009; Jacobson, 2004; Koch et al., 2009b; Rypdal et al., 2009) then calculated
31 the change in snow albedo, radiative forcing, and climate response. All of these indicate that BC in snow
32 produces warming both in the Arctic and across the northern hemisphere and that the climate efficacy
33 (change in temperature per unit forcing) of BC in snow is 2–4 times greater than that of CO_2 or other climate
34 forcers. This high efficacy occurs primarily because all of the forcing energy is deposited directly into the
35 cryosphere, whose evolution drives a positive albedo feedback on climate.

36
37 Radiative forcing by BC in the cryosphere was calculated by Jacobson (2004) and Flanner et al. (2009) using
38 a baseline snowpack which included light-absorbing soil dust, which reduces the impact of other light-
39 absorbing particles by approximately 20%. Large uncertainties persist in the distribution of dust mass and
40 absorptivity. Some model studies calculated radiative forcing due to fossil fuel and biofuel emissions only
41 (Jacobson, 2004; Rypdal et al., 2009), but others calculated the effect of all sources: fossil fuel, biofuel and
42 biomass burning (Flanner et al., 2009; Hansen and Nazarenko, 2004; Hansen et al., 2005; Koch et al.,
43 2009a). Finally, while most studies have calculated the total forcing for a given (near present-day) year,
44 Hansen et al. (2005) and Koch et al. (2009a) quantified the change in forcing from pre-industrial to present,
45 defined as 1880–2000 and 1890–1995, respectively. Key uncertainties are BC concentrations in snow and
46 ice, BC mixing state and optical properties, snow and ice area coverage and patchiness the background
47 particles already present in the snow pack, snow effective grain size and its influence on albedo reduction
48 from impurities, the masking of snow surfaces by clouds and vegetation, and the accumulation of BC at the
49 top of snowpack induced by melting and sublimation. Biases in forcing estimates arise from the model
50 assumptions cited above. We use a field survey of Arctic snow samples collected during 2005–2009
51 (Doherty et al., 2010) to adjust the model studies based on biases in Arctic BC snow concentrations. This
52 leads to a radiative effect of of $+0.04$ W m^{-2} (with a 0.01 – 0.09 W m^{-2} 90% uncertainty range) caused by
53 present-day sources of fossil fuel, biofuel, and biomass burning BC, where the range represents the
54 combination (in quadrature) of all individual uncertainties listed above except for snow patchiness and
55 masking by vegetation and clouds. This estimate includes forcing from snow on land and lying on sea-ice.
56
57

1 About 80% of this forcing is exerted on land-based snow (Flanner et al., 2007; Flanner et al., 2009). We also
2 make a crude estimate of the effect of BC within snow-free sea ice, estimating a present-day effect of +0.010
3 (0.003–0.032) $W m^{-2}$, leading to a combined (snow and sea-ice) present-day cryosphere radiative effect from
4 BC of +0.05 $W m^{-2}$. Finally, by scaling the forcing contributions from biomass burning, biofuel, and fossil
5 fuel BC emissions according to their 1750–2010 changes, we arrive at an industrial-era RF estimate of +0.04
6 (0.01–0.10) $W m^{-2}$ for the combined influence of BC in sea-ice and in snow overlying land and sea-ice. Note
7 that not all of this RF is necessarily directly anthropogenic.

8
9 Filter measurements indicate that a large fraction of the aerosol light absorption (~30–50%) in Arctic snow is
10 due to non-BC constituents (Flanner et al., 2009). Other studies show that dust is the dominant source of
11 light absorption in some continental snowpacks (Painter et al., 2010). Moreover, Hegg et al. (2010) attribute
12 most of the non-BC light absorption in Arctic snow samples to brown carbon (BrC), with crop and biomass
13 burning identified as the primary sources. These agents also contribute a positive snow radiative forcing,
14 although some of this absorption may be implicitly accounted for in the BC emission inventories applied in
15 modelling studies. As it is not clear to which extent changes in dust emissions are anthropogenically driven
16 we refrain from providing a RF best estimate for the effect of anthropogenic dust on snow and sea-ice, but it
17 is considerably smaller than the effect of BC (Flanner et al., 2009).

18
19 In summary we assess that anthropogenic absorbing aerosols (BC/BrownC) on snow/ice are responsible for a
20 positive RF of +0.04 $W m^{-2}$, with a 0.01–0.10 $W m^{-2}$ 5%–95% uncertainty range. It is important to note that
21 this forcing is 2–4 time more effective at causing global mean temperature changes than an equivalent
22 forcing from CO₂.

23 24 **7.3.6 Aerosol-Climate Feedbacks**

25 26 *7.3.6.1 Introduction*

27
28 Changes in climate parameters can modify the sources of natural aerosols and the atmospheric lifetime of
29 anthropogenic and natural aerosols, which may in turn feedback on the climate system through their
30 interactions with radiation and cloudiness and their contribution to nutrient cycling (Carslaw et al., 2010;
31 Jickells et al., 2005, see also Chapter 6). The climate drivers of changes in aerosols can be split into physical
32 changes (temperature, humidity, precipitation, soil wetness, solar radiation, wind speed, sea ice extent,
33 etc...), chemical changes (availability of oxidants) and biological changes (vegetation cover and properties,
34 plankton abundance and speciation, etc...). The response of aerosols to climate change may constitute a
35 feedback loop whereby climate processes amplify or dampen the initial perturbation. We will assess here the
36 relevance and strength of aerosol-climate feedbacks in the context of future climate change scenarios.

37 38 *7.3.6.2 Changes in Aerosol Concentrations with Climate*

39 40 *7.3.6.2.1 Sea salt and mineral dust*

41 Climate change influence atmospheric burden of sea salt by altering emissions, transport, and deposition.
42 There is no agreement among climate models about the strength of such an effect, with estimates ranging
43 from an overall 19% reduction in global sea salt burden from the present-day to year 2100 (Liao et al., 2006)
44 because of a reduction in surface wind speed and an increase in scavenging, to little sensitivity (Mahowald et
45 al., 2006a), or a sizeable increase in Bellouin et al. (2011) because of a decrease in sea ice cover despite a
46 decrease in wind speed over most of the tropical and mid-latitude oceans (Jones et al., 2007). Given that sea
47 salt particles comprise a significant fraction of CCN concentrations over the oceans, such large changes are
48 likely to feedback on climate through changes in cloud drop number (Korhonen et al., 2010b). There is little
49 understanding on how surface wind speed may change over the ocean in a warmer climate with some
50 observations suggesting an increase in wind speed over the last two decades (Young et al., 2011) and some
51 models predicting a widespread decrease in ice-free oceanic regions.

52
53 Studies of the effects of climate change on dust loadings give a wide range of results from large increases
54 (e.g., Woodward et al. (2005) find a factor of 3 increase in 2100) to moderate (–10 to –20%) increase or
55 decrease (e.g., Liao et al., 2006; Liao et al., 2009; Tegen et al., 2004) and to large decreases (e.g., Mahowald
56 and Luo (2003) and Mahowald et al. (2006b) find a 60% decrease under double CO₂ concentration). The
57 large range reflects different responses of the atmosphere and vegetation cover to climate change forcings.

1 For example, Mahowald (2007) found that the consideration of the CO₂ fertilization effect is important for
2 predicting desert response to future climate change.

3 4 7.3.6.2.2 *Sulfate, ammonium and nitrate aerosols*

5
6 The magnitude and sign of the DMS-sulfate-cloud-climate feedback loop remain uncertain despite two
7 decades of research (see Ayers and Cainey, 2007 and Carslaw et al., 2010 for a review). It is now realized
8 that the feedback could operate in numerous ways through changes in temperature, solar radiation dose,
9 mixed layer depth and nutrient recycling, sea-ice extent, wind speed, shift in marine ecosystems due to ocean
10 acidification and climate change, atmospheric processing of DMS into CCN, and no study has included all
11 the relevant effects. There is however some consistency among Earth System models to simulate a weak
12 feedback due to i) a weak sensitivity of DMS production to climate change, and ii) a weak sensitivity of
13 CCN population to changes in DMS emissions (Carslaw et al., 2010; Woodhouse et al., 2008). However
14 regional effects could be larger (Bopp et al., 2004).

15
16 In the atmosphere chemical production of sulfate increases with temperature (Aw and Kleeman, 2003;
17 Dawson et al., 2007; Kleeman, 2008), due to faster SO₂ oxidation (higher rate constants and higher oxidant
18 concentrations). Changes in O₃ and H₂O₂ with climate also influence sulfate through in-cloud sulfate
19 formation. Most studies to date predicted a reduction of 0–9% in global sulfate burden, mainly because of
20 the future increases in precipitation (Liao et al., 2006; Pye et al., 2009; Racherla and Adams, 2006; Unger et
21 al., 2006). However Rae et al. (2007) found a small increase in global sulfate burden from 2000–2100
22 because the simulated future precipitation was reduced in regions of high sulphate abundance.

23
24 Changes in temperature have a large impact on nitrate aerosol formation by shifting of aerosol
25 thermodynamic equilibrium, because more HNO₃ remains in the gas-phase in a warmer climate. There is
26 some agreement among global aerosol models that climate change will contribute to decrease nitrate
27 concentrations (Bellouin et al., 2011; Liao et al., 2006; Pye et al., 2009; Racherla and Adams, 2006) with the
28 exception of Bauer et al. (2007) who found little change in nitrate for year 2030. It should be noted however
29 that changes in precursor emissions are likely to increase nitrate concentrations in the future (Bellouin et al.,
30 2011).

31
32 Changes in sulfate and nitrate influence ammonium aerosol formation. The burden of ammonium was
33 predicted to decrease by about 5% in Pye et al. (2009) from present day to 2050 under the A1B scenario.
34 Changes in ammonium in Pye et al. (2009) are milder than those predicted by Racherla and Adams (2006)
35 and likely reflect the fact that the sulfate burden is relatively insensitive to climate change under the A1B
36 scenario.

37 38 7.3.6.2.3 *Carbonaceous aerosols*

39 Natural emissions of carbonaceous aerosols, such as wild fires and biogenic emissions are climate sensitive.
40 There is evidence that future climate change could lead to increases in the occurrence of wildfires because of
41 changes in fuel availability, readiness of the fuel to burn and ignition sources (Kloster et al., 2010; Marlon et
42 al., 2008; Mouillot et al., 2006; Pechony and Shindell, 2010). However vegetation dynamics may also play a
43 role which is not well understood. Increased fire occurrence would increase aerosol emissions, but decrease
44 BVOC emissions. This could lead to a small positive or negative radiative effect (and feedback) depending
45 on the sign of the net impact by biomass burning aerosols (Carslaw et al., 2010).

46
47 A large fraction of secondary organic carbon aerosol form from the oxidation of isoprene, sesquiterpenes and
48 monoterpenes from biogenic sources. Emissions from vegetation can increase in a warmer atmosphere,
49 everything else being constant (Guenther et al., 2006). Global aerosol models simulate an increase in
50 isoprene emissions of 22–55% by 2100 in response to T change (Heald et al., 2008; Liao et al., 2006;
51 Sanderson et al., 2003) and a change in global SOA burden of –6% to +11% through the climate-induced
52 changes in aerosol processes and removal rates (Heald et al., 2008; Liao et al., 2006; Tsigaridis and
53 Kanakidou, 2007). Increasing CO₂ concentrations are believed to inhibit BVOC emissions (Arneeth et al.,
54 2007) which could offset the T effect and adds significant uncertainty to future emissions. Future changes in
55 vegetation cover, whether they are natural and anthropogenic, also introduce large uncertainty, with either a
56 decrease (forest to cropland) or increase (some biofuels plantation) in emissions (Lathière et al., 2010). There

1 is little understanding on how the marine source of organic aerosol may change with climate,
2 notwithstanding the large range of emission estimates for the present day (Carslaw et al., 2010).

3 4 7.3.6.3 *Synthesis*

5
6 There is no robust evidence to suggest that future changes in emissions of natural aerosols (or their
7 precursors) could represent a significant climate feedback during the 21st century; the feedback factor is
8 mostly bracketed within $\pm 0.1 \text{ W m}^{-2} \text{ K}^{-1}$ (Carslaw et al., 2010). While some models simulate large changes
9 (such as for dust emissions), these simulations are also associated with large uncertainties and there is little
10 to no agreement among models. There is conflicting evidence of the strength of the feedback associated with
11 future changes in precipitation and aerosol scavenging with one study (Liao et al., 2009) showing a
12 significant positive feedback (feedback parameter of $+0.04$ to $+0.15 \text{ W m}^{-2} \text{ K}^{-1}$ on a global mean basis) and
13 other studies simulating smaller feedback of -0.02 to $-0.08 \text{ W m}^{-2} \text{ K}^{-1}$ (Bellouin et al., 2011). However such
14 feedbacks may be important for climate at the regional scale.

15 16 7.4 **Aerosol-Cloud Interactions**

17 18 7.4.1 *Introduction*

19
20 This section assesses our understanding of aerosol-cloud-precipitation interactions, emphasizing the ways in
21 which anthropogenic aerosols may be affecting the distribution of clouds and precipitation. The idea that
22 anthropogenic aerosols are changing cloudiness, and hence the planetary albedo, and thus contribute a
23 substantial radiative forcing to the climate system is a longstanding one. For these reasons aerosol-cloud
24 interactions have been addressed to varying degrees in all of the previous assessment reports.

25
26 Since the AR4, research has continued to articulate new pathways through which the aerosol may affect the
27 radiative properties of clouds, but also (and increasingly so) patterns of precipitation (Rosenfeld et al., 2008),
28 Global-scale modelling has advanced in its ability to represent a greater diversity of aerosol-cloud
29 interactions, and with greater consistency. Observational studies continue to document strong local
30 correlations between aerosol proxies and clouds or precipitation, but have become more quantitative and are
31 increasingly identifying the methodological challenges associated with such correlations. Fine-scale
32 modelling studies have begun to be used in a systematic manner, and among other things have shown how
33 turbulent mixing, cloud and regional-scale circulations may buffer the effects of aerosol perturbations
34 (Stevens and Feingold, 2009).

35
36 Aerosol-cloud interactions are highly heterogeneous and transient, which further complicates their
37 quantification. While the top-of-atmosphere radiative effect is a well-established measure of the net impact
38 of aerosol-cloud interactions on global temperatures (Lohmann and Feichter, 2005), no similar framework
39 has yet established for addressing the hydrological and/or local effects of climatic perturbations. Global-
40 mean precipitation is constrained approximately by the surface energy budget, hence an analogous
41 framework developed around the adjusted surface forcing has been suggested (Andrews et al., 2010). But
42 interest in precipitation tends to be local, where changes in horizontal energy transport within the atmosphere
43 dominate over those of the surface energy budget (Muller et al., 2011). The idea of precipitation
44 susceptibility, how the surface precipitation changes locally as a function of the aerosol burden, has been
45 developed with these interests in mind (Feingold and Siebert, 2009), and as an extension of the idea of
46 albedo susceptibility which has been used to explore the regional diversity of radiative effects stemming
47 from aerosol-cloud interactions.

48
49 The remainder of this introductory section presents an overview of the framework through which the
50 climatic effect of aerosol cloud interactions will be assessed, and highlights some of the general advances,
51 and remaining challenges that face this topic of inquiry. In the subsequent sections advancements in our
52 more specific understanding of aerosol-cloud, or aerosol-precipitation, interactions are assessed, from the
53 perspective of their overall impact on the climate system.

54 55 7.4.1.1 *Overview and Classification of Hypothesized Aerosol-Cloud Interactions*

1 Denman et al. (2007) catalogued several possible contributions to aerosol indirect effects. Given the number
2 of possible aerosol-cloud interactions, and the difficulty of isolating them individually, we see little value in
3 attempting to assess each effect in isolation especially since modelling studies suggest that the effects may
4 interact and compensate (Morrison and Grabowski, 2011; Stevens and Feingold, 2009). Instead, we group all
5 radiative consequences of aerosol-cloud interactions into two broad categories: the immediate impact on
6 radiative forcing in the absence of macrophysical changes to clouds, denoted “indirect radiative forcing”
7 (iRF), and the final result including follow-on impacts of macrophysical responses to the initial change,
8 denoted “indirect adjusted forcing” (iAF). The iRF represents the classical “Twomey” or cloud albedo effect
9 whereby greater CCN numbers increase the droplet surface area, but is extended to include ice clouds and
10 changes in the breadth of the size distribution. It is discussed further in Section 7.4.2. The iAF additionally
11 accounts for any secondary effects that result as clouds adjust to the rapid changes in their environment
12 accompanying an aerosol perturbation (Figure 7.1). Although iAF subsumes iRF, we retain an estimate of
13 iRF for continuity with prior assessments and because it is better understood than the model-dependent
14 effects determining iAF (Section 8.1.1). The iAF includes for example lifetime effects, wherein cloud
15 macrostructure adjusts to changes in cloud microstructure (Albrecht, 1989; Liou and Ou, 1989; Pincus and
16 Baker, 1994). Possible contributions to the iAF from liquid clouds are discussed in Section 7.4.3, separately
17 from those associated with adjustments by ice or mixed phase clouds, which are presented in Section 7.4.4.
18

19 *7.4.1.2 Advances and Challenges in Observing Aerosol-Cloud Interactions*

20
21 Since the AR4 the characterization of aerosol sources, sinks, and composition has continued to advance. The
22 on-going development of understanding of climate-relevant aerosol properties is reviewed in Section 7.3.3.
23

24 Progress has also been made in understanding how measurement artefacts affect retrievals of both aerosol
25 (Kahn et al., 2005; Tanré et al., 1996; Tanré et al., 1997) and cloud properties (Platnick et al., 2003) in
26 broken fields of clouds. Two key issues are that measurements classified as cloud-free often are not, and that
27 aerosol measured in the vicinity of clouds is significantly different than it would be were the cloud field, and
28 its proximate cause (high humidities), not present. The latter results from hydration effects on aerosol optical
29 properties (Charlson et al., 2007; Twohy et al., 2009), contamination by undetectable cloud fragments
30 (Koren et al., 2007) and the remote (non-columnar) effects of clouds on radiation through scattering (Wen et
31 al., 2007). Photons scattered by cloud edges can interact with clear-sky aerosol layers as far as 15 km away,
32 thereby biasing aerosol retrievals in ways that depend on the prevalence of clouds (Várnai and Marshak,
33 2009).
34

35 The use of active space-based remote sensing has also begun to address the coincidence problem, wherein
36 satellite retrievals using passive sensors are unable to distinguish aerosol layers above or below fields of
37 clouds from those intermingling with the cloud field (Anderson et al., 2005; Huffman et al., 2007; Stephens
38 et al., 2002). Spectral polarization and multi-angular measurements provide much needed information on the
39 phase function of particulate matter in the atmosphere, which can discriminate between clouds and aerosol
40 and thus improve estimates of aerosol loading and absorption (Deuzé et al., 2001; Mishchenko et al., 2007).
41 Field studies (Rauber et al., 2007; Wood et al., 2011) and laboratory investigations (e.g., Stratmann et al.,
42 2009) of cloud aerosol interactions also continue to make important contributions to our understanding of
43 how aerosols impact cloud processes. As a result our understanding of the distribution and properties of the
44 aerosol in the vicinity of clouds, continues to improve apace with an appreciation of the limits of this
45 understanding (Anderson et al., 2009).
46

47 The observational challenge of inferring causality from correlation remains a large, and limiting, one.
48 Because the aerosol is a strong function of air-mass history and origin, and is strongly influenced by cloud
49 and precipitation processes (Anderson et al., 2009; Clarke et al., 1999; Petters et al., 2006), and both are
50 affected by meteorology (Engström and Ekman, 2010), correlations between the aerosol and cloud, or
51 precipitation, cannot be taken as generally indicating a meteorological response to the aerosol. Furthermore,
52 attempts to control for other important factors (air-mass history or cloud dynamical processes) are limited by
53 a lack of understanding of cloud controlling factors in the first place (Anderson et al., 2009; Siebesma et al.,
54 2009; Stevens and Brenguier, 2009). These problems greatly undermine confidence in observationally based
55 inferences of aerosol effects on clouds and precipitation and must be considered with caution or interpreted
56 with suitable models.
57

7.4.1.3 *Advances and Challenges in Modelling Aerosol-Cloud Interactions*

Fine-scale models, capable of resolving cloud-scale circulations have greatly advanced as a tool for testing the physical mechanisms proposed to govern aerosol-cloud-precipitation interactions (Ackerman et al., 2009; vanZanten et al., 2011). Aerosol-cloud interactions in climate models have largely been introduced based on simple calculations or highly idealized models (e.g., Albrecht, 1989; Pincus and Baker, 1994; Twomey, 1977). A general finding from explicit numerical simulations of clouds is that various aerosol impact mechanisms tend to be mediated (and often buffered) by interactions across scales not included in the idealized models that gave rise to the original idea (Stevens and Feingold, 2009). Specific examples involve the interplay between the drop-size distribution and mixing processes that determine cloud macrostructure (Ackerman et al., 2004; Bretherton et al., 2007; Small et al., 2009; Stevens et al., 1998; Wood, 2007), or the dependence of precipitation development in stratiform clouds on details of the vertical structure of the cloud (Wood, 2007). As a result it is more likely than not the physical system is less sensitive to aerosol perturbations than are large-scale models.

The representation of aerosol effects in large-scale models has also advanced. Most global models now represent an increasing number of hypothesized aerosol-cloud interactions, and through comparisons to data and to other models their evaluation has greatly advanced (Quaas et al., 2009b). In particular, global models are beginning to represent effects in convective, ice and mixed-phase clouds (e.g., Lohmann, 2008) and as described below in more detail. In addition “superparameterisation” approaches (Section 7.2.3.5.2) hold promise for treating aerosol-cloud interactions more comprehensively, but are computationally very expensive and have yet to be systematically applied to this problem.

Although advances have been considerable, the challenges remain formidable. The representation of clouds in large-scale models remains primitive (Section 7.2) and even if large-scale models were able to represent clouds with greater fidelity, fine-scale modelling suggests that the outcome of an aerosol perturbation depends on the details of the interaction of clouds, turbulence, radiation and precipitation processes on a range of scales not represented by large-scale models (vanZanten et al., 2011). For this reason it is not surprising that large-scale models exhibit a range of manifestations of aerosol-cloud interactions, which limits quantitative inference (Quaas et al., 2009b). However, by examining the interactions between cloud and large-scales in the global models, and between aerosol and turbulent processes in explicit cloud simulations, some progress has been made and is likely to continue even though no single model can simulate everything.

7.4.1.4 *Combined Modelling and Observational Approaches*

Combined approaches, which attempt to maximize the respective advantage of models and data, are beginning to add to understanding of aerosol-cloud interactions. These include inversions of the observed historical record using large-scale modelling studies, but also the use of reanalysis and chemical transport models to help interpret satellite records (Chameides et al., 2002; Koren et al., 2010; Mauger and Norris, 2010), field study data to help constrain fine-scale modelling studies (Ackerman et al., 2009; vanZanten et al., 2011), or satellite climatologies to constrain large-scale modelling (Quaas et al., 2009b).

7.4.2 *Aerosol Effects on Liquid Cloud Albedo (Indirect Radiative Forcing – iRF)*

7.4.2.1 *The Physical Basis for the Indirect Forcing*

The cloud albedo effect (Twomey, 1977), which here is simply called the indirect RF or iRF, is the mechanism by which an increase in aerosol number concentration leads to an increase in liquid cloud albedo (reflectance of incoming shortwave solar radiation) by increasing the cloud droplet number concentration and hence increasing total droplet surface area, with the liquid water content and cloud geometrical thickness held fixed. Although only the change in the cloud droplet concentration is considered in the original concept of cloud albedo effect, a change in the shape of droplet size distribution (such as breadth), which is directly induced by aerosols, may also be important (e.g., Feingold et al., 1997; Liu and Daum, 2002). Cloud albedo effects may also be manifest in ice, or mixed phase clouds, but these are discussed in Section 7.4.4.

The physical basis of the indirect forcing is generally well understood with research since the AR4 generally supporting the picture that had developed at that time. Detailed in-situ aircraft observations continue to show

1 that cloud droplet concentrations observed just above the cloud base generally agree with what would be
2 predicted given the aerosol observed below the cloud base (cloud droplet closure), which is the fundamental
3 link in the cloud albedo effect (e.g., Fountoukis et al., 2007). Vertical profiles of cloud effective radius also
4 agree with those predicted by models which take into account the effect of entrainment (Lu et al., 2008),
5 although uncertainties still remain in estimating the shape of the droplet size distribution (Hsieh et al., 2009),
6 and the degree of non-adiabaticity within clouds. Multi-dimensional radiative transfer calculations have also
7 been applied to estimate cloud albedo instead of using the traditional two-stream approximation to find that
8 the latter could overestimate the albedo effect under certain conditions (Duda et al., 1996; Zuidema et al.,
9 2008).

11 7.4.2.2 *Observational Evidence for the Indirect Forcing*

12
13 There is ample observational evidence for increases in aerosol resulting in an increase in drop concentration
14 and decrease in drop size (for constant liquid water) and the main question is one of magnitude and spatial
15 extent. Based on simple metrics, there is a large range of physically plausible responses with aircraft
16 measurements (e.g., Lu et al., 2007; Lu et al., 2008; Twohy et al., 2005) tending to show stronger responses
17 than satellite-derived responses (McComiskey and Feingold, 2008; Nakajima and Michael, 2009).

18 Radiative forcing associated with the Twomey effect is impossible to observe because of the rapid
19 adjustments and further discussion is deferred to Section 7.4.3.2.

22 7.4.2.3 *Advances in Process Level Understanding*

23
24 At the heart of the albedo effect lie two fundamental issues. The first is the problem of droplet activation and
25 its sensitivity to aerosol and meteorological parameters. The primary controls on drop number concentration
26 are the aerosol number concentration (particularly at diameters > than ~ 80 nm) and updraft velocity.
27 Aerosol size distribution can play an important role under high aerosol loadings, whereas aerosol
28 composition is unimportant, except perhaps under very polluted conditions and low updraft velocities (e.g.,
29 Ervens et al., 2005). The second issue is the presence of condensed water that strongly determines how much
30 energy can be reflected; there is no indirect effect unless clouds are present. Simple arguments show that the
31 amount of reflected energy is approximately two-and-a-half times more sensitive to changes in the liquid
32 water path than to changes in drop concentration. The magnitude of the indirect effect therefore rests mostly
33 on dynamical forcing such as convective strength and entrainment that controls cloud amount, and a few key
34 aerosol parameters such as aerosol number concentration and perhaps size distribution.

35
36 While observationally-based assessments of aerosol-cloud interactions have a long history, a more recent
37 development is assessment of the ability of detailed models to reproduce the radiative fingerprints of aerosol-
38 cloud interactions (Schmidt et al., 2009). This involves comparison between measurements of fields such as
39 irradiance and comparison with the same fields calculated in finescale models that represent aerosol-cloud
40 interactions. Such approaches identify key forcing parameters (e.g., cloud-field properties, aerosol
41 hygroscopicity and absorption) and strengthen our understanding of the radiative forcing associated with
42 aerosol effects on cloud microphysics.

44 7.4.2.4 *Advances in and Insights Gained from Large-Scale Modelling Studies*

45
46 Despite the consolidation in our understanding of the physical basis of the indirect forcing there still remain
47 large uncertainties in quantification, because of the aforementioned difficulties in representing clouds and
48 aerosol-cloud interactions in climate models. Recent estimates of the indirect forcing using satellite
49 observations are systematically smaller than the AR4 estimates, which were derived from GCM calculations
50 based on parameterization of physical processes or in-situ observations (Lohmann and Lesins, 2002; Quaas
51 et al., 2008; Quaas et al., 2009b). The satellite data show lower susceptibility of cloud effective radius or
52 droplet number concentration to aerosol optical depth or number concentration as compared with in-situ
53 observations or detailed cloud parcel model calculations (McComiskey and Feingold, 2008), leading to the
54 differences in the estimate of the iRF. This result is at least partly due to scale-related averaging biases in
55 satellite retrievals (McComiskey and Feingold, 2011). It is also generally difficult to separate the iRF from
56 the cloud fast feedbacks and meteorological effects in both observations and fully coupled numerical model

1 calculations (e.g., George and Wood, 2010; Lohmann et al., 2010). Estimates of the iRF are given in Section
2 7.4.6.

3 4 **7.4.3 Adjustments in Liquid Clouds**

5 6 **7.4.3.1 The Physical Basis for Adjustments in Liquid Clouds**

7
8 The effect of the aerosol on cloud amount is more multi-faceted than its effect on albedo alone, leading
9 investigators to discuss such an effect in the plural. Such effects are often associated with changes in cloud
10 lifetime and in the past have been referred to as ‘lifetime’ effects. However this nomenclature is misleading
11 because it assumes a relationship between cloud lifetime and cloud amount. Moreover, the effect of the
12 aerosol on cloud amount may have nothing to do with cloud lifetime per se (e.g., Pincus and Baker, 1994).

13
14 The traditional view has been that indirect adjustment effects of CCN will add to the initial albedo increase
15 (iRF) by increasing cloud amounts. The chain of reasoning involves three steps. First that cloud droplet
16 concentrations depend on the number of available cloud condensation nuclei; second that precipitation
17 development is regulated by the cloud droplet concentration; and third that the development of precipitation
18 reduces cloud amount (Stevens and Feingold, 2009).

19
20 Of the three steps the first has ample support in both observations and theory, as reviewed in the previous
21 section. More problematic are the second two links in the chain of reasoning. The physical basis for an
22 unambiguous, and positive, dependence of cloud amount on the available cloud condensation nuclei is weak.
23 Although the idea that increased cloud droplet concentrations inhibit the initial development of precipitation
24 in single clouds is longstanding, it is not clear that such effects reduce the precipitation overall. Some
25 modelling studies suggest the opposite, wherein increased aerosol concentrations actually promote the
26 development of deeper clouds, thereby invigorating precipitation (Rosenfeld et al., 2008; Stevens and
27 Seifert, 2008, see also a more extensive discussion of this point in Section 7.4.5). The idea that cloud amount
28 is a decreasing function of precipitation efficiency is even less clear. Although the original studies that
29 hypothesized cloud amount effects (Albrecht, 1989; Liou and Ou, 1989) are often taken as demonstrative of
30 this point, there is limited unambiguous observational evidence (exceptions to be given below). Many
31 climate models assume such an effect *a priori*, which likely influences their forcing estimates.

32
33 A general statement of how precipitation affects cloud amount remains elusive. It appears increasingly likely
34 that cloud amount effects, which almost certainly exist locally, vary from one cloud regime to the next. As
35 such their quantification globally is a more ambitious task than originally anticipated, as it requires models to
36 correctly represent the distribution of cloud regimes evincing such effects, and knowledge of how such
37 effects manifest themselves across these regimes. Then again, because diverse effects offer the possibility of
38 compensating one another, at least globally, it seems possible that lifetime effects may be considerably less
39 important than previously thought (Stevens and Feingold, 2009).

40 41 **7.4.3.2 Observational Evidence of Adjustments in Liquid Clouds**

42
43 We discuss a sample of observations for which there is clear evidence of the aerosol interacting with clouds,
44 in the broader sense of iRF and iAF since the latter subsumes the former. The cloud albedo effect is best
45 manifested in so-called ship tracks, which are bright lines of clouds behind ships. As shown during the
46 Monterey Area Ship Track (MAST) experiment, many ship tracks are characterized by an increase in the
47 cloud droplet number concentration resulting from the increase in aerosol number concentration and an
48 absence of drizzle size drops, which leads to a decrease in the droplet radius and an increase in the cloud
49 albedo (Durkee et al., 2000). The global radiative forcing of visible ship tracks has been estimated from
50 satellite and found to be insignificant at 0.5 m W m^{-2} (Schreier et al., 2007), although there is some concern
51 that this analysis may not have identified all shiptracks. The new A-Train satellites offer the possibility to
52 study the signal of long-term degassing of low-lying volcanic aerosol on stratocumulus (Gasso, 2008) and
53 trade wind cumuli (Yuan et al., 2011). The stratocumulus respond with smaller drop sizes but ambiguous
54 changes in cloud fraction and cloud water. The trade cumuli respond with smaller droplet size, decreased
55 precipitation efficiency, increased cloud amount and higher cloud tops.

1 The development of precipitation in stratocumulus appears to be associated with more heterogeneous cloud
2 features, including breaks in the cloud coverage (Comstock et al., 2005; Sharon et al., 2006; vanZanten et al.,
3 2005). In some cases pronounced reversals in the cellular polarity become evident, where in compact regions
4 (pockets), open cellular convection is surrounded by regions of closed cellular convection. Closed cellular
5 convection in stratocumulus is characterized by a high albedo as broad regions of mesoscale ascent are
6 covered by relatively optically thick clouds and narrow regions of mesoscale descent define the cell
7 boundaries, and are characterized by optically thin clouds or even clear air. In contrast, open cellular
8 convection in stratocumulus typically has a much lower albedo; broad regions of mesoscale descent are
9 largely cloud free, and clouds are confined principally to cell boundaries in regions of mesoscale ascent. The
10 development of pockets of open cells in broad regions of closed cellular convection appears closely linked to
11 the development of precipitation (Savic-Jovicic and Stevens, 2008; Stevens et al., 2005; Wang and Feingold,
12 2009). The lack of any apparent difference in the large-scale environment of the open cells, versus the
13 surrounding closed cellular convection, implies the existence of multiple equilibria. This raises the
14 possibility that the onset of precipitation can lead to a chain of events that leads to a large-scale reduction of
15 cloudiness, in agreement with original work from Liou and Ou (1989), Albrecht (1989) and Baker and
16 Charlson (1990).

17
18 A number of hypotheses have been advanced, and explored, to explain why precipitation initially develops in
19 regions of open cells (Rosenfeld et al., 2006; Wang et al., 2010a). If broad regions of open cells were much
20 more characteristic of the pre-industrial atmosphere it would suggest that the anthropogenic aerosol has been
21 responsible for large changes in planetary albedo. Cloud resolving modelling studies suggest however that
22 even in relatively clean air in the South East Pacific, CCN concentrations are not low enough to support
23 broad regions of open cells, hence this scenario appears unlikely. Indeed, one hypothesis for the formation of
24 pockets of open cells is that they carry the imprint of airmasses in which sustained precipitation was
25 dynamically triggered thereby leading to sufficient depletion of the aerosol to sustain the observed pockets of
26 open cells (Wang et al., 2010a).

27
28 Precipitation from shallow convection, and clouds, prove difficult to observe in the trades, as the clouds tend
29 to be small, and the small footprint of many precipitating trade-wind clouds does not lend itself well to
30 space-based remote sensing techniques. [PLACEHOLDER FOR SECOND ORDER DRAFT: CloudSat
31 reference]. However unlike stratocumulus, where more precipitation favours fewer clouds, in the much
32 broader trade wind regions cloudiness tends to increase with precipitation amount, most likely because
33 processes which favour precipitation development also favour clouds (Nuijens et al., 2009) and because
34 precipitating trade cumulus tend to regenerate through colliding outflows.

35 36 7.4.3.3 *Advances in Process Level Understanding*

37
38 Since the AR4, there has been progress understanding of some basic processes relevant to cloud amount
39 effects. One basic question is how susceptible precipitation is to cloud-droplet number concentrations, and
40 by inference the available aerosol. Simple models for the conversion of cloud-water into rain-water predict
41 that the rate of this process autoconversion process scales with the square of the inverse of the droplet
42 number concentration. However theoretical work, that incorporates a fuller description of rain formation
43 processes suggests that this strongly over-estimates the sensitivity of rain formation in shallow clouds
44 (Stevens and Seifert, 2008), and that rain formation scales with a critical liquid water content that increases
45 with approximately the inverse square-root of the droplet number concentration (Kostinski, 2008; Seifert and
46 Stevens, 2010). Note that thicker, liquid clouds generate rain via accretion of cloud drops by raindrops, a
47 process that is relatively insensitive to droplet concentration, and therefore to aerosol perturbations.

48
49 A number of observational studies have likewise found that the rain-rate from stratiform clouds scales with
50 the liquid-water path of the cloud layer to the 3/2 power, and with the inverse of the droplet concentration
51 (Comstock et al., 2005; Pawlowska and Brenguier, 2003; vanZanten et al., 2005). But because the
52 observations also encapsulate the tendency of liquid water and cloud condensation nuclei to be removed by
53 rain they likely over-estimate the sensitivity of rain formation to the droplet concentration, and under-
54 estimate its sensitivity to liquid water. Some of the effects that reduce and even eliminate the sensitivity of
55 rain formation to the auto-conversion process have begun to be incorporated in parameterizations used by
56 large-scale models (Posselt and Lohmann, 2009).

1 Recent small-scale studies tend to confirm two responses of the cloud liquid water to increasing aerosol.
2 Under clean conditions when clouds are prone to precipitation, an increase in the aerosol tends to increase
3 cloud amount. Under non-precipitating conditions, clouds tend to thin in response to increasing aerosol
4 (Ackerman et al., 2004; Small et al., 2009; Xue et al., 2008). Treatment of the subtlety of these responses and
5 associated detail in small-scale cloud processes is not feasible in GCMs.

6
7 Since AR4, cloud resolving model simulation has begun to stress the importance of scale interactions when
8 addressing aerosol-cloud interactions. Large model domains (order 100 km) allow mesoscale circulations to
9 develop in response to changes in the aerosol. These dynamical responses may have a significant impact on
10 cloud morphology and radiative forcing. Examples include the dramatic changes in cloud morphology
11 associated with changes in cellular structure discussed above and the cloud-free shadows that appear
12 alongside ship tracks (Wang and Feingold, 2009). These underscore the folly of applying simplistic rules for
13 aerosol-cloud interactions.

14 7.4.3.4 *Advances in and Insights Gained from Large-Scale Modelling Studies*

15
16 Attempts to quantify cloud-mediated aerosol effects using global models suggest that lifetime effects
17 contribute between -0.3 and -1.3 W m^{-2} to the adjusted forcing from the anthropogenic aerosol (Lohmann
18 and Feichter, 2005). Because such studies usually neglect processes that may generate positive forcing
19 usually involving ice (e.g., Storelvmo et al., 2008a) and because models are tuned to preclude estimates of
20 the cloud-mediated aerosol radiative forcing that are too high (Hoose et al., 2009), it is difficult to use the
21 model-based estimates as an independent constraint. By using satellite data to rescale relations that emerge
22 from an ensemble of models several studies have argued for weaker cloud-mediated aerosol effects, and by
23 extension weaker cloud lifetime effects (Quaas et al., 2009b).

24
25
26 Regional and global models systematically misrepresent the distribution of clouds, and cloud processes,
27 especially those for shallow maritime clouds. One persistent shortcoming of global models is the tendency to
28 only treat aerosol-cloud interactions in terms of large-scale, but not convective clouds. Recent efforts to
29 consistently address both types of cloud representations represent a significant advance in large scale
30 modelling (Lohmann, 2008). Nonetheless our understanding of aerosol-cloud interactions is incomplete, and
31 what is well-understood is incompletely represented in large scale models. For these reasons, and because
32 lifetime effects depend critically on the interplay of uncertainly parameterized physical processes, global-
33 model based estimates of lifetime effects remain controversial.

34 7.4.4 *Adjustments in Cold Clouds*

35 7.4.4.1 *The Physical Basis for Adjustments in Cold Clouds*

36
37
38 In a water-saturated environment both liquid water and ice can co-exist, at least on timescales relevant for
39 cloud processes, at temperatures between 0°C and -38°C . Clouds in which both liquid water and ice particles
40 are present are referred to as mixed-phase clouds. At warmer temperatures ice rapidly melts, whereas at
41 colder temperatures liquid water will freeze homogeneously. The formation of ice in the range of
42 temperatures between 0°C and -38°C depends on heterogeneous freezing whereby a foreign medium,
43 usually insoluble aerosol particles collectively referred to as ice nuclei (IN), initiate the freezing processes.
44 Soluble matter or physiochemical transformations can hinder glaciation by depressing the freezing
45 temperature of super-cooled drops (e.g., Baker and Peter, 2008; Girard et al., 2004). The same process can
46 occur in cirrus clouds (Crawford et al., 2011) but there the lack of natural IN in the atmosphere makes
47 homogeneous freezing the preferred pathway in cirrus clouds (Kärcher and Strom, 2003). Hence
48 anthropogenic perturbations to the aerosol have the potential to affect when and where clouds become
49 glaciated. For cirrus clouds this could inhibit homogeneous nucleation. For both mixed-phase and ice clouds,
50 anthropogenic perturbations affect cloud optical properties, and can contribute to an albedo effect.

51
52
53 Because the enthalpy of liquid water is larger than that of ice, freezing is associated with a transfer of
54 enthalpy from the particles to their environment. Hence glaciation affects cloud dynamics. Moreover, in a
55 cloud consisting of supercooled liquid water the equilibrium vapour pressure will correspond to water
56 saturation, which is significantly oversaturated with respect to ice. Thus the initiation of ice in a supercooled
57 liquid cloud will cause vapour to diffuse rapidly toward ice particles at the expense of the liquid water in the

1 cloud, a process known as the Bergeron-Wegener-Findeisen process. This favours the depositional growth of
2 large ice crystals, which may sediment away from the saturated region of the atmosphere, influencing the
3 subsequent evolution of the cloud. Hence the ease with which ice forms may be influenced by anthropogenic
4 perturbations to the aerosol, and in turn may regulate cloud amount (Lohmann, 2002a; Storelvmo et al.,
5 2011; see also Section 7.2.2.4), or upper tropospheric humidity.

6
7 The types of aerosol particles that contribute to the distribution of IN apart from mineral dust are poorly
8 understood. For example, the role of biological particles acting as IN remains controversial. While such
9 particles have been found to be negligible for realistic concentrations of bacteria (Diehl and Wurzler, 2010;
10 Hoose et al., 2010a; 2010b; Phillips et al., 2009; Sesartic et al., 2011) some investigators (Ariya et al., 2009;
11 Sun et al., 2010) argue that biological particles even in low concentrations may still be important because
12 they can trigger ice multiplication. The presence of more soluble aerosol particles would make it harder to
13 form atmospheric ice homogeneously, but the primary production of insoluble aerosol particles could help
14 initiate ice through poorly known heterogeneous freezing mechanisms. If IN become coated with soluble
15 material they may also become less effective as IN (see Section 7.2.2.4, Hoose et al., 2008; Lohmann and
16 Hoose, 2009; Storelvmo et al., 2008a). Anthropogenic changes to the biosphere could conceivably also make
17 biological IN less prevalent. Our poor understanding of the climatology and life-cycle of aerosol particles
18 that can serve as IN complicates attempts to generally assess what constitutes an anthropogenic perturbation
19 to the IN population, let alone the effect of such a perturbation.

20 21 7.4.4.2 Observations of Aerosol Effects on Ice and Mixed-Phase Stratiform Clouds

22
23 Arctic mixed-phase clouds have received a great deal of attention since AR4, with major field programs
24 conducted in 2004 (Verlinde et al., 2007) and 2009 (Brock et al., 2011; Jacob et al., 2010; McFarquhar et al.,
25 2011) in addition to longterm monitoring at Barrow, Alaska (Shupe et al., 2008). Mixed-phase Arctic clouds
26 persist for extended periods of time (days and even weeks), in spite of the inherent instability of the ice-
27 water mix. We focus here on the role of the aerosol and refer to Section 7.2.1.3.3 for a discussion of
28 meteorological aspects. The subset of the aerosol that act as IN exists in concentrations of 10^{-5} to 10^{-1} cm^{-3} ,
29 i.e., only about 1 in a million particles acts as an IN. Even at such low concentrations, they have an important
30 influence on cloud persistence, with clouds tending to glaciate and disappear rapidly when IN concentrations
31 are relatively high and/or updraft velocities too small to sustain a liquid water layer. The details of the
32 heterogeneous ice-nucleation mechanism remain controversial but there is increasing evidence that ice forms
33 in Arctic stratus via the liquid phase (immersion freezing) so that the CCN population also plays an
34 important role (de Boer et al., 2011; Lance et al., 2011). If ice indeed forms via the liquid phase this
35 represents a self-regulating feedback that helps sustain the clouds: as ice forms, water is depleted, which
36 restricts further ice formation and competition for water vapour via the Bergeron-Wegener-Findeisen
37 process.

38 39 7.4.4.3 Advances in Process Level Understanding

40
41 Since the AR4 research on ice-microphysical processes has been active, to a large degree with an eye toward
42 a better representation of such processes in models. Korolev (2007) developed a theoretically based
43 parameterization of the Bergeron-Wegener-Findeisen process that has lately been employed in different
44 GCMs (Lohmann and Hoose, 2009; Storelvmo et al., 2008b). As some mixed-phase clouds have been found
45 to be long-lived, Korolev and Field (2008) derived a theoretical framework to explain this phenomenon. A
46 recent review (Morrison et al., 2012) discusses the myriad processes that create a resilient mixed-phase cloud
47 system.

48
49 An example of the level of detail associated with mixed-phase clouds that may be required for adequate
50 representation in models is that the dependence of ice particle growth by vapour diffusion depends strongly
51 on particle habit (Harrington et al., 2009) and may even have equal weight to ice nucleation mechanism vis-
52 à-vis glaciation times (Ervens et al., 2011).

53
54 For the mixed phased processes where the Bergeron-Wegener-Findeisen process makes ice-initiation
55 interesting, heterogeneous freezing parameterizations employed in cloud or larger-scale models remain
56 mostly empirical (e.g., DeMott et al., 2010; Gettelman et al., 2010; Hoose et al., 2008; Lohmann and Diehl,
57 2006; Phillips et al., 2008; Salzmann et al., 2010; Storelvmo et al., 2008a), although some recent work

1 attempts to ground the development of parameterisations in concepts derived from classical nucleation
2 theory (Hoose et al., 2010b).

3
4 Ice nucleation in cirrus clouds (at temperatures less than -35°C) depends crucially on the cloud updraft
5 velocity and hence the supersaturation with respect to ice. For homogeneous nucleation, the threshold
6 relative humidities have been parameterized using results of parcel model simulations (e.g., Barahona and
7 Nenes, 2009; Sassen and Dodd, 1988), airborne measurements in cirrus or wave clouds (Heymsfield et al.,
8 1998; Heymsfield and Miloshevich, 1995), extensions of classical homogeneous ice nucleation theory
9 (Khvorostyanov and Sassen, 1998; Khvorostyanov and Curry, 2009), and data from laboratory
10 measurements (e.g., Bertram et al., 2000; Friedman et al., 2011; Koop et al., 2000; Magee et al., 2006;
11 Mohler et al., 2003). If ice nuclei are present, then heterogeneous nucleation is the preferred freezing
12 pathway because it requires lower threshold relative humidities (or higher threshold temperatures) than
13 homogeneous nucleation. The threshold relative humidities (or temperatures) for heterogeneous nucleation
14 depend on the type and size of the involved ice nuclei (Figure 7.8 and Section 7.3.3.5).

15 7.4.4.4 *Advances in and Insights Gained from Large-Scale Modelling Studies*

16
17 Studies of the iAF that depend on ice microphysical pathways are considerably less advanced than those
18 involving only liquid clouds, but have come into increasing focus since the AR4. Penner et al. (2009)
19 obtained a rather large iAF of anthropogenic ice-forming aerosol on upper tropospheric clouds. However,
20 they ignore potential compensating effects on lower lying clouds and therefore should be regarded with
21 caution. The climate impact of anthropogenic lead-containing mineral dust particles, among the most
22 efficient ice-forming substances, has been investigated. In the extreme scenario in which 100% of ice-
23 forming mineral dust particles in cirrus clouds contained lead, up to 0.8 W m^{-2} more long-wave radiation
24 was emitted to space as compared to pure mineral dust particles (Cziczo et al., 2009b).

25
26 Since the AR4 a number of studies have contributed to our ability to quantify aerosol effects on cirrus.
27 Because such clouds usually only involve ice-phase microphysical processes, the physical pathways are
28 somewhat simpler. BC can impact background cirrus by affecting ice nucleation properties but its effect
29 remains uncertain (Kärcher et al., 2007). There is some evidence of a statistically significant impact on cirrus
30 coverage (Hendricks et al., 2005). However, Liu et al. (2009) examined their role in radiative forcing of
31 cirrus clouds and found it to be very small.

32 7.4.5 *Aerosol-Cloud Microphysical Effects on Precipitating Systems*

33 7.4.5.1 *The Physical Basis for Aerosol Effects on Precipitating Convection*

34
35
36 Deep convective clouds arise in response to the differential heating of the surface versus the atmosphere,
37 largely as a result of radiative processes. This differential heating leads to the development of convective
38 instabilities. The enthalpy of vaporization, and the much smaller enthalpy of fusion, which is released
39 through the development of precipitation, is a primary mechanism for transporting enthalpy from the surface
40 to the atmosphere and hence consuming the column instability. Through their effect on the microphysical
41 development of clouds, for instance whether or not precipitation sized particles are readily formed, or if ice
42 is initiated, the aerosol may modify the vertical distribution of condensate, and thus modify cloud and
43 precipitation development.

44
45
46 Microphysically, the suppression in drop size in response to an aerosol perturbation is expected to suppress
47 drop growth and precipitation formation and favour enhanced water loading in the upper portions of the
48 cloud, which may limit cloud development, but favour the formation of convective downdrafts through more
49 ready mixing with the environment. However, the lack of precipitation may allow clouds to grow deeper and
50 help generate more precipitation (Nuijens et al., 2009). Then again, the cold pools associated with
51 precipitation may be crucial for secondary precipitation (Lee et al., 2010; Matheou et al., 2011). As a result,
52 whether more efficient precipitation production fosters or limits cloud development remains very uncertain.

53
54
55 Similar arguments can be extended to the ice phase. Once the cloud passes the freezing level, the effect of
56 the aerosol on ice initiation becomes important, with the ensuing complications discussed in the previous
57 section. If, however, changes in the anthropogenic aerosol make the initiation of ice less effective the

1 development of precipitation may be further retarded (Rosenfeld and Woodley, 2001), and vice versa. By
2 delaying the initiation of ice the additional enthalpy of fusion will be introduced higher in the convective
3 updraft, thereby suppressing the development of clouds initially, but promoting their development when they
4 reach the level where ice forms. Although this is a rather small effect, relative to the tenfold larger enthalpy
5 release from ongoing condensation, it has nonetheless been hypothesized to have a critical effect on cloud
6 development (Andreae et al., 2004; Rosenfeld et al., 2008).

7
8 Finally, by determining whether liquid, mixed, or ice-phase processes are dominant, perturbations to the
9 aerosol may also affect the distribution of precipitation. Studies in the southwestern Amazon region during
10 the transition from dry to wet seasons (Martins et al., 2009) invoke such mechanisms to explain why higher
11 CCN concentrations are associated with less frequent low-to-moderate rainfall rates and more frequent high
12 rainfall rates.

13
14 To date, the issue of aerosol effects on precipitation remains an open question (Levin and Cotton, 2009).
15 There are many uncertainties related to the physical mechanisms involved and to the observational and
16 numerical tools (e.g., Khain, 2009).

17 18 7.4.5.2 *Observations of Aerosol Effects on Precipitating Systems*

19
20 Numerous observational studies have examined the links between aerosol particles and deep convective
21 cloud properties, seeking to find robust associations between the aerosol and cloud properties that can be
22 interpreted to be a result of aerosol effects on clouds. The availability of satellite data with global coverage,
23 ever finer footprints, and a richer palette of spectral, angular, and polarization information has improved the
24 retrieval of aerosol and cloud properties, and continues to advance the field, with the major strides since the
25 AR4 coming from the use of active remote sensing, angular, and vertically, resolved data.

26
27 The analysis of satellite data, and *in situ* measurements, shows that an increase in aerosol loading is
28 associated on average with smaller particles, taller invigorated convective clouds with larger cloud fraction
29 and more extensive ice portions. These associations are found over the tropical Atlantic (Jenkins et al., 2008;
30 Koren et al., 2005), Europe (Devasthale et al., 2005), North and South America (Andreae et al., 2004; Bell et
31 al., 2008; Koren et al., 2008; Lin et al., 2006; Lindsey and Fromm 2008) and appear for all types of aerosol
32 particles: biomass burning smoke, urban/industrial aerosol and desert dust. These more invigorated clouds
33 would be expected to create more precipitation (Rosenfeld et al., 2008). For example an analysis of satellite
34 data over the entire Brazilian Amazon during the dry, biomass burning, season found that elevated aerosol
35 loading was associated with increased rainfall amounts and a shift to higher rainfall rates (Lin et al., 2006).
36 Likewise changes in rainfall during the week over the south-eastern portion of the United States found an
37 increase in rainfall for afternoon storms during the midweek compared to the weekend (Bell et al., 2008).

38
39 Observational studies examining the aerosol effect on precipitation of mixed phase clouds, often however
40 report rain suppression in polluted atmospheres. Satellite data measured at various geographical locations
41 suggest suppressed precipitation associated with polluted clouds (Jiang et al., 2008). Investigation of
42 orographic clouds showed a reduction in the annual precipitation over topographical barriers downwind of
43 major urban areas (Givati and Rosenfeld, 2004; Jirak and Cotton, 2006). Other studies showed no response
44 of orographic rain to pollution (Halfon et al., 2009).

45
46 However, whether these observations should be interpreted as an effect of the aerosol on clouds, vice versa,
47 or due to a third factor remains controversial. Causal pathways have been proposed that would be consistent
48 with all of the hypotheses (Rosenfeld et al., 2008; Stevens and Feingold, 2009). What is clear is that a
49 consistent picture has yet to emerge, perhaps because different pathways are active in different cloud
50 regimes, or at different stages in the life-cycle of cloud systems (Koren et al., 2008; Stevens and Seifert,
51 2008).

52 53 7.4.5.3 *Advances in Process Level Understanding*

54
55 Modelling studies suggest that the thermodynamic environment in which the clouds grow is an important
56 factor in the determination of the aerosol effect on the ground precipitation (Khain et al., 2005; Lynn et al.,
57 2005; Tao et al., 2007). For clouds developing in dry unstable air, there is a decrease in the accumulated

1 precipitation with an increase in aerosol loading. For deep maritime clouds modelling studies suggest that
2 aerosol perturbations can lead to an increase in precipitation. The important environmental factors are
3 humidity, that influences the evaporation of cloud liquid (and sublimation of ice), and wind shear that can
4 modulate the entrainment of dry air into clouds and the transport of cloud liquid into unsaturated areas (Fan
5 et al., 2009).

6
7 Weekly cycles in aerosol properties and precipitation have emerged to tackle this problem. While all studies
8 support a weekly cycle in aerosol properties, the results for weekly cycles in precipitation are conflicting
9 (Barnet et al., 2009; Bäumer et al., 2008; Hendricks Franssen et al., 2009; Sanchez-Lorenzo et al., 2008;
10 Stjern 2011).

11
12 Recently the tools of numerical weather prediction have been applied to this problem. Looking at summer
13 season forecasts using a cloud-resolving regional model the question has been posed as to whether changes
14 to the aerosol systematically affect precipitation over large regions. Little to no systematic effect of the
15 aerosol could be documented (Seifert et al., 2011).

16
17 Cloud-resolving modelling by Lynn et al. (2007) suggests that aerosol perturbations will result in
18 precipitation being displaced to the leeward side of the mountain, with the impact of the aerosol being
19 strongest under drier conditions and weaker horizontal winds. Regional studies confirm a reduction of
20 precipitation on the windward side of a mountain barrier and a tendency for the precipitation to shift
21 downstream to the leeward side but the magnitude of this depends on the importance of the ice phase in these
22 orographic clouds (Muhlbauer and Lohmann, 2009; Zubler et al., 2011).

23
24 As computational resources have increased it has become increasingly clear that some aerosol effects appear
25 as transients in short duration simulations and that long (multi-day) simulations are necessary to get a more
26 complete picture. Moreover the need to consider cloud systems, as opposed to individual clouds, when
27 attempting to establish aerosol influences must be emphasised (Morrison and Grabowski, 2011). Parallels
28 can be drawn with shallow systems in which mesoscale organization triggered by aerosol perturbations
29 amplifies the response beyond what one might have expected from microphysical considerations alone (see
30 Section 7.4.3.3). The large-scale constraint on surface precipitation makes it much more likely that one
31 might get changes in the frequency and spatial distribution of rain, than changes in the total amounts.

32 33 *7.4.5.4 Advances in and Insights Gained from Large-Scale Modelling Studies*

34
35 Fast feedbacks associated with the aerosol indirect effects do not cause much change in precipitation if an
36 average over a big enough domain is considered. Slow feedbacks through aerosol-induced changes in surface
37 temperature/surface energy budget and changes in circulation can cause regional and global changes in
38 precipitation. These latter effects have been estimated in AR4. Here the decrease in the global annual mean
39 shortwave radiation at the surface since pre-industrial times due to scattering and absorbing aerosols
40 amounted to -2.3 W m^{-2} with a range between -1.3 to -3.3 W m^{-2} (Denman et al., 2007). The associated
41 change in the global mean precipitation amounts to between 0 and $-0.13 \text{ mm day}^{-1}$ (Denman et al., 2007).

42
43 Studies with a climate model coupled to an advanced representation of the aerosol, wherein convection is
44 parameterized, also find little evidence of a weekly cycle in precipitation (Quaas et al., 2009a).

45 46 *7.4.5.5 Large-Scale Convective Systems*

47
48 Several studies since the AR4 highlight the possibility of aerosol effects on Atlantic hurricane activity, either
49 by altering radiative heating or through microphysical effects on clouds. Year-to-year variations in hurricane
50 activity and prevalence of dusty Saharan air layers (SAL) are significantly anticorrelated (Dunion and
51 Velden, 2004; Evan et al., 2006). Periods of prominent SAL activity correlate with reduced sea surface
52 temperatures, which would tend to suppress strong storm activity (Lau and Kim, 2007); dust also absorbs
53 sunlight, thus warming the troposphere and further decreasing instability (Jury and Santiago, 2010; Wong et
54 al., 2009). Evan et al., (2008) estimated that one third of the increase in the hurricane power dissipation
55 index during the preceding 25 years was statistically attributable to the absence of the SAL. Since the SAL is
56 warm and dry, it would be expected to suppress convection even in the absence of any dust (Sun et al., 2008;
57 Wong and Dessler, 2005), which complicates any attribution of observed behaviour to dust. Moreover, Folz

1 and McPhaden (2008) showed that the correlation between Saharan dust and a cooler sea surface was mainly
2 because both were caused by stronger winds, rather than dust causing the cooling. Thus while it is likely that
3 dust variations have to some extent affected tropical Atlantic sea surface temperature trends and therefore
4 Atlantic hurricane activity, it remains unclear how important this is.

5 6 **7.4.6 Synthesis of Aerosol Effects**

7
8 There are different ways to report the iRF and the iAF at the top-of-the-atmosphere since pre-industrial
9 times. In Figure 7.13a we show estimates of the change in the net radiation if possible. If that estimate is not
10 available the next options, taken in this order, are the change in the net shortwave radiation, the change in the
11 net cloud forcing and last, the change in shortwave cloud forcing. Given that there is practically no longwave
12 signal associated with the iRF and there are no changes in the clear-sky, all estimates of the iRF are
13 comparable. However, for the iAF when fast feedbacks are included, there can be changes in the clear-sky
14 and the longwave radiation, especially if aerosol effects on mixed-phase and ice clouds are considered or if
15 local circulations respond rapidly to changes in the shortwave forcing.

16
17 Ensemble-averaged global-mean model estimates of the iRF have remained rather constant over time (Figure
18 7.13a) and amount to roughly -1 W m^{-2} . This estimate is obtained from the average over all published
19 estimates, treating each of them as equal (one vote per model per paper). The -1 W m^{-2} estimate is slightly
20 stronger than the estimate of the indirect forcing in AR4 where a different weighting procedure was used
21 (Forster et al., 2007). If the iRF studies from GCMs are divided into those published prior to TAR (1993–
22 2000), between TAR and AR4 (2001–2006) and since 2007, the median indirect forcing remains within 0.15
23 W m^{-2} . It is most negative and exhibits the largest variability between 2001 and 2006 (Figure 7.13b). This
24 increase in variability reflects the increase in complexity with which aerosol-cloud interactions are
25 simulated. Whereas early models used offline three-dimensional sulphate fields, state-of-the art GCMs have
26 their own aerosol schemes and consider sea salt, mineral dust and carbonaceous aerosols in addition to
27 sulphate. There does not seem to be a systematic tendency for models that use a parameterization based on
28 cloud parcel models instead of empirical relationships between the aerosol mass/number concentration with
29 the cloud droplet number concentration to have a larger or smaller indirect aerosol effect. Sensitivity studies
30 did show that the iRF is larger if the background aerosol concentration is low (Chen and Penner, 2005) as
31 this increases the cloud susceptibility. As shown by Storelvmo et al. (2009) different empirical relationships
32 that are used to bypass cloud activation can cause a difference of 1.3 W m^{-2} in the iRF. The iRF also depends
33 strongly on the assumed minimum cloud droplet concentration because that determines the susceptibility of
34 the cloud (Hoose et al., 2009). The iRF is smallest (-0.4 W m^{-2}) if model data are rescaled to conform with
35 observational constraints.

36
37 In response to the aerosol, there are multiple possible adjustments, such as changes to the cloud lifetime
38 (cloud lifetime or second indirect aerosol effect), reduction in cloud cover due to absorption of solar
39 radiation by BC or other absorbing aerosols (semi-direct effect) and aerosol effects on mixed-phase, ice and
40 convective clouds (Denman et al., 2007) that are included in the iAF. The iAF amounts to -1.5 W m^{-2} if
41 either changes in cloud lifetime alone or changes in cloud lifetime together with the direct and semi-direct
42 aerosol effects are included in GCMs. Note that GCMs that use autoconversion rates of cloud droplets to
43 form rain drops which depend inversely on the cloud droplet number concentration build in a cloud lifetime
44 effect. In small-scale studies this does not lead to an increase in lifetime because small droplets also
45 evaporate more readily (Jiang et al., 2006) but rapid timescale processes of this kind are not represented in
46 GCMs (Lohmann and Feichter, 2005). The iAF tends to be smaller if changes to the cloud droplet size
47 distribution (dispersion) are considered (e.g., Rotstayn and Liu, 2005) or if a prognostic equation for
48 precipitation is introduced (Posselt and Lohmann, 2009) because that shifts the emphasis from the
49 autoconversion rate to the accretion between cloud droplet and rain drops in better agreement with
50 observations (Wood, 2005).

51
52 iAF is considerably smaller if aerosol effects on mixed-phase clouds are included in addition to the above
53 mentioned adjustments in liquid clouds. The GCM average amounts to -1.1 W m^{-2} , only slightly larger than
54 the iRF implying that the rapid adjustments (and inclusion of the direct aerosol effect) almost cancel each
55 other. The spread between the different studies in the liquid+mixed category depends on the frequency of
56 glaciation of supercooled clouds. If more IN are available in a polluted climate, supercooled clouds glaciate
57 more readily and precipitate (see Section 7.4.4.1). In these cases an additional cooling stems from more

1 longwave radiation being emitted to space. If on the contrary IN become coated with soluble material and
2 become less efficient, supercooled clouds remain longer in the atmosphere, which enhances the iAF of liquid
3 clouds but leads to a small positive longwave effect that slightly reduces the shortwave cooling.

4
5 The iAF increases substantially in magnitude if aerosol particles are also allowed to change convective
6 clouds. However, the uncertainty associated with these estimates is the largest and also it is unclear if
7 convective clouds are characterized in sufficient detail in GCMs to warrant such estimates. As in the case of
8 the iRF, rescaling model based estimates of the iAF, so as to match constraints from satellite retrievals, also
9 reduces their magnitude (from -1.1 to -0.7 W m^{-2} , Figure 7.13a).

10
11 A complementary approach to estimate the iAF is to infer it as a residual using the observed temperature
12 record over land, and estimates of the ocean heat uptake and the evolution of greenhouse gas and solar
13 radiative forcing (Anderson et al., 2003; Hegerl et al., 2007). These approaches are called inverse estimates.
14 They normally involve models of intermediate complexity. The only inverse study that obtained the iAF
15 (Knutti et al., 2002) bracketed the iAF to be between 0 and -1.2 W m^{-2} . All estimates of iAF that involve
16 satellite data fit into this range as do the mean values of the different iAF groups. An inverse estimate that is
17 obtained purely from an energy balance perspective limits the iAF (including the direct effect plus other
18 unknown residuals that are assumed to be small) since 1950 to be between -0.7 to -1.5 W m^{-2} (Murphy et
19 al., 2009). The 5–95% confidence interval of all inverse estimates of the iAF is -0.4 to -1.3 W m^{-2} (Figure
20 7.13b). Again, all estimates of iRF that involve satellite data fit into this range as does the GCM average of
21 the iAF that includes mixed-phase clouds. However, all other iAF averages exceed the 95% confidence
22 interval of the inverse estimates. This is likely reflects limitations in our ability to parameterize clouds,
23 aerosols, and aerosol-cloud interactions in GCMs.

24
25 Because GCMs tend to include negative forcings but not positive ones they tend to produce larger forcings
26 than small-scale studies that include compensating processes and than inferred from observations (e.g., ship
27 track studies or pure GCM estimates of the iAF and iRF vs. those that include satellite data). Therefore we
28 use the GCM estimates of the iRF (average of iRF-TAR, iRF-AR4 and iRF-AR5 in Figure 7.13) as
29 providing the lower bounds of -1 and -1.5 W m^{-2} . The lower bound of the iAF is obtained from the average
30 of iAF-liquid-AR4 and iAF-liquid-AR5 as most of the other studies also include the direct effect. It amounts
31 to -1.5 W m^{-2} . The upper bounds have been put at the smallest GCM estimates of -0.1 W m^{-2} and 0 W m^{-2}
32 for the iRF and iAF, respectively, because of the indications that GCMs overestimate the forcings and that
33 very small forcing values cannot be ruled out. The studies that take satellite data into account therefore arrive
34 at much smaller forcings with a median of -0.4 W m^{-2} and -0.7 W m^{-2} for the iRF and iAF, respectively and
35 an upper bound for the iRF of -0.2 W m^{-2} . We regard the median values of the studies including satellite
36 data as a plausible but yet more uncertain estimate of the lower bound. Based on these arguments, we assess
37 the iRF and iAF as follows: iRF is very likely between -1 and -0.1 W m^{-2} , the lower bound being based on
38 GCMs, and likely between -0.4 and -0.2 W m^{-2} , the lower bound being based on studies that take satellite
39 data into account. Following the same line of argumentation, iAF is very likely between -1.5 and 0 W m^{-2}
40 and likely between -0.7 and -0.2 W m^{-2} .

41 42 **[INSERT FIGURE 7.13a HERE]**

43 **Figure 7.13a:** Model, satellite and inverse estimates of the iRF and the iAF since 1993. For each paper the best estimate
44 per model is shown as a plus-sign. If multiple estimates or uncertainties are given in a paper, the ranges are shown as
45 vertical lines bounded by diamonds. The thin horizontal lines denote the average of the respective group and the width
46 of the coloured box denotes its standard deviation. The iRF studies from GCMs are divided into those published prior to
47 TAR: iRF-TAR (Boucher and Lohmann 1995; Chuang et al., 1997; Feichter et al., 1997; Jones et al., 1994; Kaufman
48 and Chou 1993; Kiehl et al., 2000; Lohmann and Feichter 1997; Lohmann et al., 2000; Rotstayn 1999), between TAR
49 and AR4: iRF-AR4 (Chen and Penner 2005; Chuang et al., 2002; Ghan et al., 2001; Hansen et al., 2005; Jones et al.,
50 2001; Kristjansson 2002; Ming et al., 2005; Penner et al., 2006; Quaas and Boucher 2005; Quaas et al., 2004; Rotstayn
51 and Penner 2001; Rotstayn and Liu 2003; Suzuki et al., 2004; Takemura et al., 2005; Williams et al., 2001) and since
52 2007: iRF-AR5 (Barahona et al., 2011; Bellouin et al., 2011; Haerter et al., 2009; Kvalevag and Myhre 2007; Lohmann
53 et al., 2007; Lohmann et al., 2010; Penner et al., 2011; Rotstayn and Liu 2009; Storelvmo 2011; Storelvmo et al., 2009;
54 Wang and Penner 2009). iAF studies on liquid clouds that include the cloud albedo and cloud lifetime effect are also
55 divided into those published until 2006: iAF-liquid-AR4 (Easter et al., 2004; Ghan et al., 2001; Johns et al., 2006; Jones
56 et al., 2001; Kristjansson 2002; Kristjansson et al., 2005; Lohmann 2002b; Lohmann and Feichter 1997; Lohmann et
57 al., 2000; Menon et al., 2002; Ming et al., 2005; Peng and Lohmann 2003; Penner et al., 2003; Penner et al., 2006;
58 Quaas et al., 2006; Rotstayn 1999; Rotstayn and Penner 2001; Rotstayn and Liu 2005; Storelvmo et al., 2006;
59 Takemura et al., 2005; Williams et al., 2001) and since 2007: iAF-liquid-AR5 (Chen et al., 2010; Ghan et al., 2011b;

Hoose et al., 2009; Kirkevåg et al., 2008; Makkonen et al., 2011; Menon and DelGenio 2007; Ming et al., 2007b; Penner et al., 2011; Quaas et al., 2009b; Rotstajn and Liu 2009; Storelvmo et al., 2008a); iRF and iAF estimates that involve satellite data are shown in pink: iRF-satellites (Dufresne et al., 2005; Lebsock et al., 2008; Quaas and Boucher 2005; Quaas et al., 2008; Quaas et al., 2009b; Storelvmo et al., 2009) and iAF-satellites (Lohmann and Lesins 2002; Quaas et al., 2006; Quaas et al., 2009b; Sekiguchi et al., 2003), inverse estimates for the iRF and iAF are shown in turquoise: iRF-inverse (Knutti et al., 2002) and iAF-inverse (Anderson et al., 2003; Andronova and Schlesinger 2001; Church et al., 2011; Forest et al., 2006; Forest et al., 2002; Gregory et al., 2002; Hansen et al., 2011; Harvey and Kaufmann 2002; Huber and Knutti 2011; Libardoni and Forest 2011; Murphy et al., 2009; Shindell and Faluvegi 2009; Stott et al., 2006) in turquoise. iAF studies that include the direct and semi-direct effect in lilac: iAF-liquid+dir+SD (Ghan et al., 2011a; Lohmann and Feichter 2001; Lohmann et al., 2007; Posselt and Lohmann 2008; Posselt and Lohmann 2009; Quaas et al., 2004; Quaas et al., 2006; Quaas et al., 2009b; Rotstajn et al., 2007; Salzmann et al., 2010), those that additionally consider aerosol effects on mixed-phase clouds in purple: iAF-liquid+mixed (Hoose et al., 2008; Hoose et al., 2010b; Jacobson 2006; Lohmann 2004; Lohmann and Diehl 2006; Lohmann and Hoose 2009; Lohmann and Ferrachat 2010; Salzmann et al., 2010; Storelvmo et al., 2008a; Storelvmo et al., 2008b), and those that treat aerosol effects in stratiform and convective clouds in green: iAF-liquid+conv (Koch et al., 2009a; Lohmann 2008; Menon and Rotstajn 2006; Menon and DelGenio 2007; Unger et al., 2009; Wang et al., 2011b). For the inverse estimates no best estimate is shown and the turquoise colour box denotes the average of the lower and upper bounds of these studies, respectively.

[INSERT FIGURE 7.13b HERE]

Figure 7.13b: Box plots of model, satellite and inverse estimates of the IF and the AIF since 1993 for the same groups of estimates as in Figure 7.13a provided at least 6 estimates are available. Displayed are the averages (red stars), median values (blue lines), 33% and 67% percentiles (box boundaries) and 5% and 95% percentiles (ends of vertical lines) except for the inverse estimates, which is an expert assessment of the combined estimate of multiple inverse estimates.

Finally Table 7.3 provides values of the aerosol AF as diagnosed in simulations of the CMIP5 models which provide climate projections in this report. [PLACEHOLDER FOR SECOND ORDER DRAFT: to discuss differences between our best estimates and CMIP5]

Table 7.3: Estimates of aerosol AF (in $W m^{-2}$) in the CMIP5 models. The AF are estimated from fixed-SST experiments using the atmosphere-only version of the models listed. Different models include different aerosol effects. [PLACEHOLDER FOR SECOND ORDER DRAFT: table will be updated as data become available on the CMIP5 archive.]

Modelling Group	CCCma	CSIRO-QCCCE	IPSL	MOHC	NCC	MPI-M
Model	CanESM2	CSIRO-Mk3-6-0	IPSL-CM5A-LR	HadGEM2-A	NorESM1-M	
Anthropogenic sulfate aerosol		-1.10	-0.71	-1.16		
All anthropogenic aerosol	-0.87	-1.41		-1.22	-0.99	-0.35

7.4.7 Impact of Cosmic Rays on Aerosols and Clouds

A high solar activity leads to a more complex magnetic configuration of the heliosphere, which reduces the flux of galactic cosmic rays (GCR) in the Earth's atmosphere. It has been hypothesised that a lower flux of GCR would modify cloudiness in a way that would amplify the warming effect expected from an increase in solar activity. There have been many studies aiming to test this hypothesis since AR4, which fall in two categories: i) studies that seek to establish a causal relationship between cosmic rays and aerosols/clouds by looking at correlations between the two quantities on timescales of days to decades, and ii) studies that test through observations or modelling one of the physical mechanisms that have been put forward. We assess these two categories of studies in the next two sections.

7.4.7.1 Correlations Between Cosmic Rays and Properties of Aerosols and Clouds

Many empirical relationships or correlations have been reported between GCR or cosmogenic isotope archives and some aspects of the climate system, such as SSTs in the Pacific Ocean (Meehl et al., 2009), some reconstruction of past climate (Kirkby, 2007) or tree rings (Dengel et al., 2009). We focus here on observed relationships between GCR and aerosol- and cloud-properties. Such relationships have focused on

1 decadal variations in GCR induced by the 11-year solar cycle, shorter variations associated with the quasi-
2 periodic oscillation in solar activity centred on 1.68 years or sporadic variations associated with so-called
3 Forbush decrease events that happen on timescales of days. It should be noted however that such correlations
4 could arise for reasons related to changes in atmospheric heating and circulation, rather than an impact of
5 cosmic rays.

6
7 Some studies have shown co-variation between GCR and low-level cloud cover using global satellite data
8 over periods of typically 5–10 years (e.g., Marsh and Svensmark, 2000; Svensmark and Friis-Christensen,
9 1997). Such correlations have not proved to be very robust when extending the time period under
10 consideration (Agee et al., 2011), restricting the analysis to particular cloud types (Kernthaler et al., 1999) or
11 locations (Udelhofen and Cess, 2001; Usoskin and Kovaltsov, 2008). Some authors have attributed the
12 purported correlations to other factors such as El Niño (Farrar, 2000) and artefacts of the satellite data due to
13 the solar cycle cannot be ruled out (Pallé, 2005). However statistically significant correlations have been
14 found in some locations. Harrison and Stephenson (2006) examined the relationship between diffuse
15 radiation and cosmic rays at the surface for some UK sites between 1951 and 2000. They found that the 87%
16 of days with the highest cosmic ray flux had a $19\pm 4\%$ higher chance of being overcast and the diffuse
17 fraction in incoming surface solar radiation increases by $2\pm 0.3\%$. Moreover Forbush reductions in cosmic
18 rays corresponded to a simultaneous decrease in diffuse fraction. Similarly Harrison (2008) found a unique
19 1.68 year cosmic ray periodicity in surface radiation for two different UK sites between 1978 and 1990,
20 which they believe is associated with variations in cloudiness. These cloud responses were found to occur
21 within a day (Harrison and Ambaum, 2010). However, large reduction events in cosmic rays did not occur
22 frequently enough in the record to generate robust statistics. Svensmark et al. (2009) found large global
23 reductions in the aerosol Angström exponent from AERONET, liquid water path from SSM/I, and cloud
24 cover from MODIS and ISCCP after large Forbush decreases with a lag of 5 to 9 days. This finding has been
25 questioned by Laken et al. (2009). The study by Kristjansson et al. (2008) suggests a weaker impact of
26 Forbush decrease event on clouds over the Southern Ocean. Moreover the 5–9 day lag found by Svensmark
27 et al. (2009) does not match the rapid response observed in Harrison and Stephenson (2006) and Harrison
28 and Ambaum (2010). Further the studies of Calogovic et al. (2010) and Kristjansson et al. (2008) did not
29 find a global cloud effect. Laken et al. (2010) and Rohs et al. (2010) found a very weak but significant
30 positive correlation between GCR and high- and mid-altitude clouds. A problem with all these studies is that
31 very few large Forbush decrease events have occurred during the satellite era (~6) and the statistics are
32 sensitive to how the Forbush events are selected (Laken et al., 2009). Finally it should be noted that Kulmala
33 et al. (2010) found no connection between GCR and new particle formation over a solar cycle (1996–2008);
34 however the measurements of new particle formation were restricted to one surface station in Finland and
35 may not be representative of new particle formation in the free troposphere.

36 37 7.4.7.2 *Physical Mechanisms Linking Cosmic Rays to Cloudiness*

38
39 Several physical mechanisms have been put forward to explain the possible link between GCR and
40 cloudiness. The most widely studied of these is the “ion-aerosol clear air” mechanism, in which atmospheric
41 ions produced by GCR facilitate aerosol nucleation and growth with a further impact on CCN concentrations
42 and cloud properties (Carslaw et al., 2002; Usoskin and Kovaltsov, 2008). The variability of atmospheric
43 ionization rates due to GCR changes can be considered relatively well quantified (Bazilevskaya et al., 2008),
44 whereas resulting changes in aerosol nucleation rates are very poorly known (Enghoff and Svensmark, 2008;
45 Kazil et al., 2008). The Cosmics Leaving Outdoor Droplets (CLOUD) experiment at CERN has been
46 designed to simulate conditions close to the real atmosphere and is currently the only laboratory experiment
47 on ion-induced nucleation in which it has been possible to isolate the role of ions produced by GCRs (Kirkby
48 et al., 2011). The CLOUD experiment indicates that GCR-induced ionization enhances water–sulphuric acid
49 nucleation in the middle and upper troposphere, but is very unlikely to give a significant contribution to
50 nucleation taking place in the continental boundary layer. Field measurements support qualitatively this view
51 but cannot provide any firm conclusion on the role of ions because of the scarcity and other limitations of
52 free-troposphere measurements (Arnold, 2006; Mirme et al., 2010) and difficulties in separating GCR-
53 induced nucleation from other nucleation pathways in continental boundary layers (Hirsikko et al., 2011;
54 Manninen et al., 2010). Regardless of the exact aerosol formation mechanism, a significant fraction of CCN
55 in the boundary layer may originate from aerosol particles nucleated in the free troposphere (Merikanto et
56 al., 2009).

1 Observational evidence for particle nucleation in the cloudy marine boundary layer is relatively rare with
2 some notable exceptions (e.g., Hegg et al., 1990; Petters et al., 2006). Nucleation (neutral and charged)
3 appears to require special conditions, namely low ambient aerosol, high DMS, photochemically produced
4 OH (to oxidise DMS to SO₂), relatively high RH and relatively low temperature. The previously discussed
5 open-cellular cloud structure appears to provide adequate conditions for nucleation. Updrafts in open cell
6 walls supply DMS produced at the ocean surface to an ultra-clean layer near the top of the boundary layer
7 that has been scavenged of aerosol by precipitation. High actinic flux because of the presence of adjacent
8 clouds produces OH, which oxidises the DMS to SO₂ and H₂SO₄. Petters et al. (2006) observed new particle
9 formation within open cells and more recently Kazil et al. (2011) modeled the formation of particles via
10 neutral and charged H₂SO₄/H₂O nucleation in a mesoscale cloud-resolving model representing the aerosol
11 lifecycle. The relative importance of surface aerosol production, entrainment from the free troposphere, and
12 nucleation are shown depend on factors such as surface wind-speed, entrainment rates, and just how
13 favourable ambient conditions are for nucleation. To date, no process-level models have looked at the role of
14 the phase of the solar cycle, via its influence on nucleation rates, on this balance of aerosol sources and their
15 importance for maintaining boundary layer clouds.

16
17 Our understanding on the “ion-aerosol clear air” as a whole relies on a few model investigations that
18 simulate GCR changes over a solar cycle (Kazil et al., 2006; Pierce and Adams, 2009a; Snow-Kropla et al.,
19 2011) or during strong Forbush decreases (Bondo et al., 2010; Snow-Kropla et al., 2011). Although all
20 model studies found a detectable connection between GCR variations and either CCN changes or column
21 aerosol properties, the response appears to be too weak to cause a significant radiative effect because of the
22 low sensitivity of CCN concentrations to the nucleation caused by GCR.

23
24 A second mechanism has been proposed by which ionization in the atmosphere may have an impact on
25 clouds. GCR ionization modulates the fair-weather current in the global electrical circuit and it has been
26 hypothesised that droplet charging could modify supersaturation and temperature at the cloud base. Harrison
27 and Ambaum (2010) found some observational evidence of this with a small reduction in downward
28 longwave radiation associated with variations in surface current density, but the evidence remains very low.

30 7.4.7.3 *Synthesis*

31
32 In summary, there is evidence from laboratory, field and modelling studies that ionization from cosmic rays
33 may enhance aerosol nucleation in the free troposphere. However there is medium evidence and high
34 agreement that the cosmic ray-ionization mechanism is too weak to influence global concentrations of CCN
35 or their change over the last century or during a solar cycle in a climatically-significant way. Moreover it
36 should be noted that one study infers no trend in cosmic ray intensity over the last 50 years (McCracken and
37 Beer 2007).

39 7.5 Solar Radiation Management and Related Techniques

41 7.5.1 *Introduction*

42
43 Geoengineering is a term often used to describe the deliberate large scale intervention in the Earth system to
44 counter undesirable impacts of climate change on the planet (e.g., Keith, 2000). One class of geoengineering
45 methods is based upon manipulating the energy budget of the planet. Solar Radiation Management (SRM)
46 methods aim to achieve a planetary cooling by reducing the amount of solar energy absorbed by the climate
47 system. They are discussed here because some of these methods involve clouds and/or aerosols. A related
48 technique is also discussed that seeks to deliberately decrease the greenhouse effect in the climate system by
49 altering high-level cloudiness. The other class of geoengineering methods known as Carbon Dioxide
50 Reduction (CDR) is discussed in Chapter 6.

51
52 This section restricts its assessment to a “physical climate” perspective of methods published in the scientific
53 peer-reviewed literature that appear to influence components of the energy budget by at least a few tenths of
54 a W m⁻² in the global mean and for which a plausible technology exists. Cost, implementation and
55 governance issues are beyond the scope of this section. [PLACEHOLDER FOR SECOND ORDER DRAFT:
56 reference to WGIII AR5 (tbc)]. Geo-engineering techniques were not discussed in previous WGI
57 assessments but were mentioned in WGII (Klein et al., 2007) and WGIII (Barker et al., 2007) AR4.

1
2 Virtually all research on SRM has followed one of two paths: i) theoretical and modelling studies and ii) a
3 search for analogues to SRM, and the impact of those phenomena to the planet (e.g., large volcanic eruptions
4 in the stratosphere such as Pinatubo, ship tracks, inadvertent climate modification from anthropogenic
5 aerosols, etc). The scientific tools required to explore SRM methods and their impact on the planet are
6 essentially the same as those needed to understand and predict the impacts of other forcing agents. The
7 predicted climate changes in response to SRM are subject to the same limitations and uncertainties as
8 predictions of future climate change. We assess geoengineering from two points of view: i) issues
9 surrounding the production of the radiative forcing and ii) the climate response and other impacts generated.
10 As with other types of climate forcing, there are serious limitations in our capability to predict regional
11 responses of the climate system to SRM.

12 13 **7.5.2 Idealised Experiments**

14
15 Some aspects of SRM methods can be explored in an idealised way by artificially reducing the solar constant
16 in a climate model. This results in a global cooling, albeit with a climate efficacy generally smaller than 1,
17 and a reduction in the global-mean precipitation. Earlier (Bala et al., 2008) and more recent (GeoMIP,
18 Kravitz et al., 2011) model experiments where the RF by CO₂ is exactly balanced by a reduction in the solar
19 constant show some residual surface temperature changes, especially at high latitudes, and a reduction in the
20 global-mean precipitation which can be explained by arguments on the energy budget of the atmosphere
21 (Allen and Ingram, 2002; Andrews et al., 2010; Held and Soden, 2006). Figure 7.14 shows the annual-mean
22 temperature and precipitation changes produced by experiment G1 of the Geoengineering Model
23 Intercomparison Project for a subset of 4 models (GeoMIP, Kravitz et al., 2011).

24
25 [PLACEHOLDER FOR SECOND ORDER DRAFT: a more complete discussion of impacts on T, P, sea-ice
26 and soil moisture in the GeoMIP models.]

27 28 **[INSERT FIGURE 7.14 HERE]**

29 **Figure 7.14:** Multi-model mean of the residual surface temperature and precipitation changes from GeoMIP
30 simulations with a simultaneous fourfold increase in CO₂ and a reduction in solar forcing which has been adjusted in
31 each model to maintain the top of atmosphere net flux imbalance within $\pm 0.1 \text{ W m}^{-2}$ (Kravitz et al., 2011).

32
33 Idealized experiments have also been conducted where solar radiation is reduced only over the ocean (e.g., to
34 mimic the effects of marine cloud seeding or increased sea foam). Bala et al. (2010) suggested that although
35 global-mean precipitation might decrease, precipitation over land might actually increase, because of
36 increased gradients in RF between land and ocean. Other studies have explored albedo changes over land
37 (e.g., to represent a surface albedo increase through plant albedo and desert regions) and over particular
38 latitudinal bands (Caldeira and Wood, 2008) or regions (Irvine et al., 2010; Irvine et al., 2011).

39 40 **7.5.3 Stratospheric Aerosols**

41
42 Observations in the aftermath of major volcanic eruptions like Pinatubo demonstrate that increasing
43 stratospheric aerosols will cool the planet and it has been suggested that global warming might be
44 deliberately countered with a continuous release of sulphur species (Budyko, 1974; Crutzen, 2006) to mimic
45 this. Most of the research on stratospheric aerosol SRM to date has explored the possibility of forming
46 sulphuric acid aerosols by injecting sulphur containing gases in the stratosphere, although using BC
47 (Crutzen, 2006; Keith, 2010) or metal oxides (Keith, 2010) has also been suggested.

48
49 There are many subtleties to SRM by stratospheric aerosols (Rasch et al., 2008b). The evolving size of the
50 particles in the stratosphere has profound effects on the viability of the strategy, with impacts on radiative
51 forcing by unit of injected mass, stratospheric ozone chemistry, and climate response. Initial modelling
52 studies recognized this fact but prescribed the aerosol size, assuming it would range between small sizes
53 characteristic of background conditions and the larger sizes observed soon after a major volcanic eruption
54 (Rasch et al., 2008a). Heckendorn et al. (2009) found that particle size from a continuous injection of gases
55 oxidizing to sulphuric acid particles could be very inefficient, because much of the sulphuric acid would
56 condense on particles already present from earlier SRM emissions. Particles would be likely to grow larger,
57 become less efficient per unit mass at scattering energy back to space, and sediment quicker out of the

1 stratosphere as the injection flux increases. They found that at least a four times higher injection rate would
2 be required to double the forcing for the scenario they explored. Pierce et al. (2010) have suggested a way
3 around this by introducing a source of sulphuric acid gas that immediately condenses to particles, bypassing
4 some coagulation and deposition processes that lead to particle growth from gaseous sulphur precursors.
5

6 Several modelling studies suggest that it would be possible to stabilize global average surface temperature at
7 least through a doubling of CO₂ concentrations using stratospheric sulphate aerosol with many signatures
8 and planetary consequences that are similar to those found in the idealized studies, i.e., a residual warming at
9 high latitudes (assuming a more or less homogeneous distribution of stratospheric aerosols) and a reduction
10 in the global-mean precipitation.
11

12 Dispersion and lifetime of the aerosol dispersed in the stratosphere is a strong function of height and latitude
13 of the injection, with high latitude and lower injection altitudes being less effective. Local injections of
14 aerosol precursors at high latitudes will produce aerosol extending over sizable fraction of a hemisphere.
15 Early studies (Jones et al., 2010a; Rasch et al., 2008b; Robock et al., 2008) used somewhat different
16 experimental protocols and found significant disagreement in regional responses to stratospheric aerosols. It
17 is unclear whether differences in the regional responses are due to the experimental protocol, or to model
18 differences. [PLACEHOLDER FOR SECOND ORDER DRAFT: GeoMIP update]
19

20 Observations also show other impacts from volcanic eruptions like Pinatubo. There are measurable effects
21 on the hydrologic cycle (Trenberth and Dai, 2007) similar to those found in idealised experiments, impacts
22 on stratospheric ozone, and the ratio of direct to diffuse sunlight reaching the Earth's surface. These effects
23 are also expected to occur from geoengineering with sulphate aerosols (see Rasch et al., 2008b and
24 references therein). Tilmes et al., (2009) used a model with a well resolved middle atmosphere and
25 stratospheric chemistry to explore responses to stratospheric aerosol SRM and found discernable shifts in
26 tropopause altitude (lifting by 1 km), and changes in ozone abundance (depletion at high latitudes and
27 increases in the tropics). This change in ozone might have discernable impacts on UV light reaching the
28 surface, although some earlier calculations suggest that there is some degree of compensation between
29 increases in UV associated with ozone depletion and decreases associated with attenuation by the aerosols
30 themselves (Vogelmann et al., 1992). A decrease in direct radiation and increase in diffuse radiation reaching
31 the Earth's surface may increase photosynthesis in terrestrial ecosystems (Mercado et al., 2009; see Chapter
32 6) and impact some systems that exploit renewable solar energy [WGII Chapter xx].
33

34 **7.5.4 Cloud Brightening**

35
36 Boundary layer clouds act to cool the planet, and relatively small changes in cloud albedo, lifetime, or areal
37 extent can have profound effects on the energy budget of the planet (e.g., Slingo, 1990). Latham (1990)
38 suggested that it might be possible to deliberately increase cloud albedo as a mechanism for countering
39 global warming by introducing additional sea salt particles into the marine boundary layer, to act as CCN,
40 "brightening" clouds through the aerosol-cloud indirect mechanisms described in Section 7.4. The idea has
41 been examined using models at various scales (cloud parcel models, large eddy simulations, and climate
42 models (e.g., Latham et al., 2008). Examples of cloud brightening include shiptracks produced in marine
43 stratocumulus clouds by emissions of particles from freighters and changes in trade cumulus cloud properties
44 produced by a relatively weak but continuous volcanic eruption of SO₂ (Yuan et al., 2011).
45

46 Changing cloud morphology (e.g., from open to closed cells) or changing low-liquid water clouds to high
47 liquid water clouds have the potential to create large radiative forcings. Our current understanding suggests
48 that marine stratocumulus clouds are an optimal cloud type for brightening because of their relatively low
49 values of CDNC and the longer lifetime of sea-salt particles in non-precipitating environments. However
50 these clouds occupy a relatively small fraction of the planet and large RF (30–100 W m⁻²) would be required
51 locally to produce globally-averaged changes of the order of 1–5 W m⁻². Studies cited in Section 7.4
52 highlight the importance of the details of aerosol-cloud interactions in influencing cloud albedo and lifetime
53 producing very large uncertainties about the viability of cloud brightening for SRM. Wang et al. (2011a)
54 explored the sensitivity of marine stratocumulus in various meteorological regimes and levels of background
55 aerosol amounts to aerosol injection strategies using a cloud system resolving model. That work
56 demonstrated very strong interactions between aerosol distribution, precipitation and cloud morphology and
57 differing responses in each scenario. Korhonen et al. (2010a) showed that "competition effects" between the

1 geoengineering aerosol and ambient aerosol could be important, with the ambient and SRM aerosol
2 populations competing for liquid water, sometimes reducing, rather than enhancing albedo in some
3 circumstances.

4
5 [PLACEHOLDER FOR SECOND ORDER DRAFT: contribution of geo-engineered sea-salt particles to the
6 direct effect to be discussed].

7
8 Climate model studies (Jones et al., 2009; Latham et al., 2008; Rasch et al., 2009) that assumed
9 geoengineering would directly influence cloud drop number changed cloud albedo and produced global
10 average RF as negative as -5 W m^{-2} . These studies also indicated some changes in regional precipitation
11 patterns, although the sign and amplitude of the changes differed between studies.

12 **7.5.5 Surface Albedo Changes**

13
14 It has also been suggested that planetary albedo can be increased by local changes to the albedo of urban
15 areas, croplands, grasslands, deserts and the ocean surfaces.

16
17 Rosenfeld et al. (1998) proposed to increase the albedo of urban areas as a way to improve air quality and
18 make cooling-energy savings in buildings especially during summertime. Hamwey (2007) estimated the
19 potential RF from whitening roofs and pavements at -0.17 W m^{-2} but more recent estimates accounting for
20 more realistic artificial surface area per capita and appropriate atmospheric radiative transfer suggest
21 significantly less negative values (Akbari et al., 2009; Lenton and Vaughan 2009; Oleson et al., 2010).

22
23 Hamwey (2007) further suggested that increasing the albedo of the world's grassland (meaning open
24 shrubland, grassland and savannah) by replacing native species by other natural or bioengineered species.
25 Their RF estimate of -0.59 W m^{-2} (assuming the grassland albedo can be increased by 25% from an average
26 value of 0.17) has been revised to -0.5 W m^{-2} by Lenton and Vaughan (2009) when accounting for the
27 atmospheric absorption of the radiation reflected by the surface. Ridgwell et al. (2009) and Doughty et al.
28 (2011) extended the concept of increased surface albedo to croplands and found the maximum effect over
29 summertime mid-latitudes (e.g., 0.25 K per 0.01 increase in surface albedo in regions north of 30°N).
30 Ridgwell et al. (2009) estimated a global-mean surface cooling of 0.11 K for a +0.04 increase in cropland
31 albedo. Both studies pointed out to potential feedbacks in low-latitude regions with a reduction in soil
32 moisture, cloud cover and precipitation. The potential for increasing crop and grassland albedo across a wide
33 variety of species remains unproven.

34
35 Irvine et al. (2011) tested the impact of increasing desert albedo up to 0.80 in the HadCM3 model. This
36 cooled surface temperature by -1.1 K (versus -0.22 and -0.11 K for their largest crop and urban albedo
37 change). They also simulate significant land precipitation changes, with large reduction in rainfall over the
38 Indian and Sahel regions.

39
40 The low albedo of ocean surfaces and large areal extent mean only a small increase in albedo could be
41 sufficient to offset several W m^{-2} of RF by greenhouse gases. Engineering techniques (Evans et al., 2010;
42 Seitz, 2011) have been proposed to increase the fraction of the oceans covered with foam because of its large
43 albedo (Whitlock et al., 1982). However ocean foam is short-lived and artificial foam would somehow have
44 to be engineered to last longer. Neither the extent of foam generation and persistence required for a
45 significant climate impact nor the impact of artificial foam on the world's ocean (including ocean biology,
46 air-sea fluxes of latent heat, sensible heat and trace gases, and surface emissivity) have been assessed in the
47 peer-reviewed literature.

48 **7.5.6 Cirrus Thinning**

49
50 Cirrus clouds affect both outgoing longwave radiation (OLR) and absorbed solar radiation. Thin high cirrus
51 above 300 hPa affects OLR more than incoming solar energy and thereby contribute to warming the climate
52 (see Section 7.2). Reducing the coverage or longwave opacity of these clouds would therefore contribute a
53 negative RF. Cirrus cloud coverage is sensitive to the ice fall speed which depends on ice crystal size. By
54 increasing ice crystal size in the coldest cirrus ice crystals could fall out and reduce the overall coverage
55 (Mitchell and Finnegan, 2009). Although an aerosol cloud seeding mechanism has been proposed to increase
56
57

1 crystal size, cirrus nucleation processes are not yet well enough understood to provide a firm basis to this
2 method (Section 7.4).

3
4
5 **[START FAQ 7.1 HERE]**

6
7 **FAQ 7.1: How do Aerosols Affect Climate and Climate Change?**

8
9 **[INSERT FAQ 7.1, FIGURE 1 HERE]**

10 **FAQ 7.1, Figure 1:** Overview of aerosol direct and indirect effects on climate.

11
12 It is believed that man-made variations in atmospheric aerosols are responsible for a cooling which have
13 partially masked the warming from man-made greenhouse gases.

14
15 Atmospheric aerosols are small particles suspended in the atmosphere with a typical lifetime of 1–2 weeks in
16 the troposphere and 1–2 years in the stratosphere. There are many types of aerosols, which can be of natural
17 (e.g., dust, sea-salt, some biogenic compounds) or anthropogenic (e.g., sulphates, soot, biomass burning
18 aerosols) origin. Atmospheric aerosols exhibit large variations in size, shape and chemical composition.
19 Changes in the climate can be caused either by emissions of anthropogenic aerosols and their precursors, or
20 by changes in natural aerosols which themselves respond to other changes in the climate system (e.g.,
21 increase in dust due to a regional drying).

22
23 Aerosols affect climate in multiple ways. First they scatter and absorb sunlight which modifies the planet
24 radiative balance, an effect known as the *aerosol direct effect* (see FAQ.7.1, Figure 1). Aerosol scattering
25 generally results in a more reflective planet and a cooler global climate, while absorption results in a less
26 reflective planet and a warmer climate. Sulphate aerosols from fossil fuel burning are especially important in
27 scattering, while soot from some combustion sources is an important absorber. The balance between cooling
28 and warming depends on the aerosol properties and the environmental conditions. Many observational and
29 modelling studies have been done to quantify the global direct effect from anthropogenic and natural
30 aerosols. While these remain uncertain, studies have consistently indicated that the direct effect from
31 anthropogenic aerosols has been to cool the planet relative to what would otherwise have occurred. One of
32 the remaining uncertainties comes from aerosol absorption, which is more difficult to measure than
33 scattering and induces a specific cloud response (known as the *semi-direct effect*).

34
35 Since aerosols are distributed unevenly in the atmosphere, they can heat and cool the climate system in
36 patterns that can drive subtle changes to the weather, affecting cloud or rainfall amounts. These effects are
37 complex and hard to predict with current models, but several studies suggest significant effects on
38 precipitation in certain regions.

39
40 Aerosols also serve as condensation and freezing sites for cloud droplet and ice particle formation (see
41 FAQ.7.1, Figure 1). While it might seem that more condensation nuclei would increase the amount of low
42 clouds, cloud formation is largely limited by dynamical processes so that the net effect on clouds of more
43 aerosols is quite subtle and remains uncertain. A robust result is that more aerosols tend to produce liquid
44 clouds with more numerous and smaller particles, everything else being equal. This and other impacts on
45 clouds alter their reflectivity, producing what are called *aerosol indirect effects* on climate. Indirect effects
46 can arise through many pathways, particularly in ice or mixed liquid and ice clouds where phase changes are
47 sensitive to aerosols. Quantifying the overall impact is understandably more difficult than for the direct
48 effect, but again available studies generally indicate that the net indirect effect of anthropogenic aerosols has
49 been to further cool the climate system over the industrial period, enhancing their direct effect.

50
51 Because of their short lifetime, the abundance of aerosols and their climate effects have varied over time in
52 rough concert with anthropogenic emissions of aerosols and their gaseous precursors, and variations in
53 natural sources. Since anthropogenic emissions have increased substantially over the industrial period, this
54 has very likely counteracted some of the warming that would otherwise have occurred from increased
55 concentrations of long-lived greenhouse gases. Aerosols from volcanic eruptions such as those of the El
56 Chichón and Pinatubo have also caused sporadic cooling periods. Trends in anthropogenic aerosol emissions
57 over the last couple of decades have varied regionally (e.g., decreased emissions in industrialised countries,

1 increased emissions in developing countries) and it is difficult to assess whether the global impact has been
2 to cool or warm the planet over the recent period. It is very likely, however, that emissions of anthropogenic
3 aerosols will ultimately decrease. When this happens, decreasing aerosol emissions will begin to augment
4 greenhouse-gas induced warming.

5
6 Some studies have hypothesised that climate change could feed back on the lifecycle of atmospheric
7 aerosols, including natural aerosols such as sulphate, sea salt or biogenic aerosols. However there is
8 contradicting evidence so far whether this could be a large effect at the global scale over the coming century.
9 Moreover the sign of such a feedback is not known and could vary regionally.

10
11 **[END FAQ 7.1 HERE]**

12
13
14 **[START FAQ 7.2 HERE]**

15 **FAQ 7.2: How do Clouds Affect Climate and Climate Change?**

16
17
18 The importance of potential changes in cloudiness for the problem of climate change has been recognized as
19 a key factor since the 1970s. Clouds affect the climate system in a variety of ways. They produce
20 precipitation (rain and snow) that is necessary for life. They strongly affect the flows of both solar and
21 infrared radiation through the atmosphere. Finally, they are intimately associated with powerful vertical
22 motions that can carry air from near the surface to the upper troposphere in less than an hour. The strong
23 vertical currents carry energy, moisture, momentum, and various chemical constituents, including aerosols.
24 Each of the various cloud processes has the potential to change as the climate state evolves. Any change in a
25 cloud process that is caused by a climate change and in turn influences climate represents a cloud-climate
26 feedback.

27
28 Cloud feedbacks are of intense interest in the context of anthropogenic climate change. Many types of
29 possible cloud-climate feedbacks have been identified. Broadly speaking, they would occur through changes
30 in cloud amount, cloud top-height, and/or cloud optical properties. We still are not sure what types of cloud
31 feedbacks will actually occur and how significant they will be for climate change. Nevertheless, all of the
32 models used for the fourth IPCC Assessment produced either a positive or near-neutral cloud feedback. The
33 differences in cloud feedbacks among the models strongly influenced their differences in climate sensitivity.

34
35 Low clouds reflect a lot of solar radiation back to space, but have only a weak effect on the infrared radiation
36 emitted by the Earth. As a result, they tend to cool the Earth, in the present climate. In a future climate
37 warmed by increasing greenhouse gases, an increase in low cloud amount would increase the cooling, and so
38 could reduce the warming. On the other hand, a decrease in low-cloud amount would increase the warming.

39
40 Conversely, high cold clouds such as cirrus clouds are often somewhat transparent, so they do not reflect as
41 much solar radiation, but they can still absorb the infrared radiation coming from the Earth's surface, leading
42 to a warming near the cloud-base level and reducing the energy Earth loses to space. They therefore tend to
43 warm the Earth as a whole. An increase in high cloud amount would tend to enhance greenhouse warming,
44 while a decrease would tend to reduce it. Even if the high-cloud amount remained the same, high clouds
45 could produce a positive feedback as the surface warms up, because they would prevent the extra infrared
46 energy emitted by the warmer surface from leaving the climate system.

47
48 The amount of sunlight a typical cloud would reflect in a warmer climate could be different for many
49 reasons. As an example, a warmer climate may see more clouds made of liquid drops, and fewer made of ice
50 crystals. That could lead to a change in the overall amount of light reflected. Clouds are also affected in
51 many ways by aerosols (see FAQ 7.1), which may have caused significant past changes (or may cause future
52 changes) in cloud reflectivity independent of any caused by climate change. Subtle changes in wind patterns
53 associated with transient or longer-term climate changes would also likely affect clouds.

54
55 In a climate change, there can be many different changes in the geographical patterns and seasonal
56 distributions of both high and low clouds. The net cloud feedback results from the combined effect of these
57 various changes.

1
2 For decades, climate scientists have been using observations to study how clouds change with the daily
3 weather, with the seasonal cycle, and with year-to-year changes such as those associated with El Niño. We
4 have also been working to improve the simulation of clouds in climate models. Many current models predict
5 a moderately positive net cloud feedback, in which both low and high clouds feed back positively. Work
6 continues to further evaluate and refine these results.

7
8 [PLACEHOLDER FOR SECOND ORDER DRAFT: addition of comment about confidence in current
9 models is being considered]

10
11 The net feedback from clouds on global climate, if any, will almost surely result from the net effect of many
12 diverse regional changes. This makes predicting the cloud feedback very difficult. While it would be
13 desirable to infer this long-term cloud feedback somehow from observations, there is no way to do this that
14 is broadly accepted as valid. To predict cloud phenomena comprehensively requires a global climate model;
15 these models produce cloud fields that roughly resemble those observed, but are far from perfect. Models
16 vary in how they predict clouds will change in a warmer climate, but so far no model has predicted changes
17 in clouds so large that they significantly limit global warming, and nearly all models predict that cloud
18 changes will actually amplify global warming.

19
20 **[END FAQ 7.2 HERE]**

21
22
23 **[START FAQ 7.3 HERE]**

24 25 **FAQ 7.3: Could Geoengineering Counteract Climate Change and What Side-Effects Might Occur?**

26
27 There are two different categories of geoengineering methods which are usually referred to as Solar
28 Radiation Management (SRM, assessed in Chapter 7) and Carbon Dioxide Removal (CDR, assessed in
29 Chapter 6). A less technical name for SRM is Sunlight Reflection Management. We discuss these in turn.

30 31 **Carbon dioxide removal methods**

32
33 By definition, CDR methods seek to accelerate the removal of CO₂ from the atmosphere and store it in land,
34 ocean or geological reservoirs. Afforestation/reforestation, carbon sequestration in soils, bioenergy
35 associated with carbon capture and storage, ocean fertilization, accelerated weathering of silicate and carbonate
36 rocks and CO₂ air capture using chemical methods are some of the proposed CDR methods (see FAQ.7.3,
37 Figure 1).

38
39 **[INSERT FAQ 7.3, FIGURE 1 HERE]**

40 **FAQ 7.3, Figure 1: Overview of carbon dioxide removal methods.**

41
42 CDR methods rely primarily on natural carbon cycle processes, either biological or chemical: enhanced
43 biological production by photosynthesis on (1) land and (2) oceans, (3) accelerated chemical weathering
44 reactions over (3) land and oceans and (4) enhanced solubility pump in the oceans. Direct air capture is an
45 exception which relies on artificial chemical methods to remove CO₂ directly from air. Once captured, CO₂
46 would be stored within land and ocean reservoirs or geological formations. Land storage occurs in organic
47 form but storage in oceans and geological formations is in inorganic forms.

48
49 To have a noticeable climate effect, CDR schemes should be able to remove several PgC per year from the
50 atmosphere over several decades in this century. Important scientific considerations include the storage
51 capacity and permanence of the reservoirs, and potential adverse side effects. CDR methods cause a
52 “rebound effect”: when carbon is stored in one reservoir, the concentration gradient between the atmosphere
53 and carbon reservoirs is reduced and thereby the subsequent inherent rate of removal of CO₂ from the
54 atmosphere by natural reservoirs is reduced or could even be reversed.

55
56 In general, CDR methods are believed to be relatively low risk in terms of unintended climatic side effects
57 because they counter the root cause by reducing atmospheric carbon dioxide concentrations. CDR schemes

1 also reduce direct consequences of high CO₂ levels including surface ocean acidification. However, proposed
2 CDR methods have limited potential to rapidly decrease the atmospheric concentration of CO₂. The large
3 thermal inertia of climate system need to be also considered: many components of the earth system may
4 continue to respond for decades or centuries to the original increases in CO₂ even after CDR is applied.
5 Therefore, decreases in surface temperature would lag CDR-induced decreases in atmospheric CO₂
6 concentrations.

7
8 There are some potential climate or environmental effects from CDR methods. Some examples of the side
9 effects are: 1) removal of atmospheric CO₂ would lead to a temporary acceleration in the global water cycle.
10 2) Implementation of CDR methods could lead to reduced plant productivity when compared to the elevated
11 level expected with high CO₂ concentration. 3) Large scale biological production over land could have
12 climate consequences by altering the surface characteristics such as surface reflectivity and
13 evapotranspiration. For instance, many modelling studies have shown that afforestation in seasonally snow
14 covered regions could accelerate global warming. 4) In the case of ocean fertilization, the utilization of
15 macronutrients such as nitrogen and phosphate in the fertilized region can lead to a decrease in production
16 "downstream" from the fertilized region. 5) Ocean iron fertilization could acidify the deep ocean by storing
17 more dissolved inorganic carbon there. 6) Enhanced biological production over oceans could potentially lead
18 to expanded regions with low oxygen concentration, increased production of N₂O and CH₄, and possible
19 disruptions to marine ecosystems.

20
21 While the rate of removal of CO₂ through accelerated weathering and direct air capture methods are limited
22 primarily by cost, energy and environmental constraints, those methods involving biological processes
23 operate much more slowly with estimates of maximum physical potential for atmospheric CO₂ removal for
24 each of the more effective biological CDR strategies to be on the order of 100 Gt C (~ 50 ppmv of CO₂) over
25 a century, with similar limitations from cost, energy and environmental issues.

26 27 **Solar radiation management methods**

28
29 The average temperature of the planet is controlled by the amount of sunlight absorbed by the Earth's
30 atmosphere and surface, and the degree to which gases and clouds in the atmosphere hinder the escape of the
31 energy back to space. If less incoming sunlight reaches the surface (because of an increase in the reflectivity
32 of the planet) or if it becomes easier for energy to escape (because of a decrease in heat trapping gases or
33 some types of clouds), the average surface temperature will decrease.

34
35 Geoengineering methods relying on managing the Earth's radiative budget are based on this fundamental
36 physical principle. These methods seek to increase the reflectivity of the planet by making the atmosphere,
37 clouds or the surface more reflective or by suppressing cirrus clouds that hinder the escape of energy from
38 the Earth system (see FAQ.7.3, Figure 2).

39 40 **[INSERT FAQ 7.3, FIGURE 2 HERE]**

41 **FAQ 7.3, Figure 2:** Overview of solar radiation management methods.

42
43 Basic physics tells us that if either of these changes is successful, the planet will cool. The picture is
44 complicated, however, because of the multiple complex physical processes that govern the interactions
45 between the flow of energy, the atmospheric circulation, weather and the resulting climate. While the
46 average surface temperature of the planet responds to the energy budget in a rather straightforward way, the
47 temperature at any particular location and time is influenced by many other factors. It is expected that any
48 particular geoengineering technique will cool some regions more than others: the locations where radiation
49 management cools the planet need not correspond to the locations and times where greenhouse gases
50 produce a warming. For example, a change in the amount of sunlight via radiation management will operate
51 only during daytime, but changes in greenhouse gases affect heating rates during both day and night. This
52 inexact compensation will have some influence on the diurnal cycle of surface temperature at any given
53 location, even if the average surface temperature is unchanged. This is a simple example of inexact
54 compensation but other subtle changes may also occur.

55
56 Climate is much more than surface temperature, however; it is also characterized by precipitation patterns,
57 the distribution of snowpack and sea-ice, and the frequency of occurrence of extreme events, just to name a

1 few. Both models and theory show that compensating an increased greenhouse effect with an increased
2 planetary reflectivity will not maintain both the average surface temperature and the average precipitation
3 rate. Regional changes in heating/cooling are expected to affect local precipitation rates, and other aspects of
4 climate. The imprecise compensation in regional and global climate patterns make it unlikely that solar
5 radiation management will produce a future climate that is “just like” the one we experience today, or have
6 experienced in the past. The residual climate changes from inexact compensation may increase as the
7 geoengineering effort is scaled up.

8
9 In addition, solar radiation management techniques may also have other side effects. For example, radiation
10 management by stratospheric sulphate aerosols can produce stratospheric ozone depletion, and changes in
11 the ratio of direct to diffuse sunlight reaching the surface that can affect terrestrial ecosystems. Moreover,
12 radiation management will not have any impact on ocean acidification, which is driven by the atmospheric
13 CO₂ concentration. A key unanswered question is whether or not the benefits of radiation management
14 outweigh the associated risks, in light of the expected residual impacts.

15
16 **[END FAQ 7.3 HERE]**
17

References

- 1 **References**
- 2
- 3 Abbatt, J. P. D., S. Benz, D. J. Cziczo, Z. Kanji, U. Lohmann, and O. Mohler, 2006: Solid ammonium sulfate
- 4 aerosols as ice nuclei: A pathway for cirrus cloud formation. *Science*, **313**, 1770-1773.
- 5 Abdul-Razzak, H., and S. Ghan, 2000: A parameterization of aerosol activation 2. Multiple aerosol types. *J.*
- 6 *Geophys. Res.*, **105**, 6837-6844.
- 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 aerosol climate forcing. *Nature*, **432**, 1014-1017.
- 9 Ackerman, A. S., and Coauthors, 2009: Large-eddy simulations of a drizzling, stratocumulus-topped marine
- 10 boundary layer. *Monthly Weather Review*, **137**, 1083-1110.
- 11 Adachi, K., S. H. Chung, and P. R. Buseck, 2010: Shapes of soot aerosol particles and implications for their
- 12 effects on climate. *Journal of Geophysical Research*, **115**, D15206.
- 13 Adams, P. J., J. H. Seinfeld, D. Koch, L. Mickley, and D. Jacob, 2001: General circulation model assessment
- 14 of direct radiative forcing by the sulfate-nitrate-ammonium-water inorganic aerosol system. *Journal of*
- 15 *Geophysical Research*, **106**, 1097-1111.
- 16 Agee, E. M., K. Kiefer, and E. Cornett, 2011: Relationship of lower troposphere cloud cover and cosmic
- 17 rays: An updated perspective. *Journal of Climate*, **early online release**.
- 18 Ahern, H. E., K. A. Walsh, T. C. J. Hill, and B. F. Moffett, 2007: Fluorescent pseudomonads isolated from
- 19 Hebridean cloud and rain water produce biosurfactants but do not cause ice nucleation.
- 20 *Biogeosciences*, **4**, 115-124.
- 21 Akbari, H., S. Menon, and A. Rosenfeld, 2009: Global cooling: increasing world-wide urban albedos to
- 22 offset CO₂. *Climatic Change*, **94**, 275-286.
- 23 Albrecht, B. A., 1989: Aerosols, cloud microphysics, and fractional cloudiness. *Science*, **245**, 1227-1230.
- 24 Alexander, D. T. L., P. A. Crozier, and J. R. Anderson, 2008: Brown carbon spheres in East Asian outflow
- 25 and their optical properties. *Science*, **321**, 833-836.
- 26 Allan, R. P., and B. J. Soden, 2008: Atmospheric warming and the amplification of precipitation extremes.
- 27 *Science*, **321**, 1481-1484.
- 28 Allan, R. P., B. J. Soden, V. O. John, W. Ingram, and P. Good, 2010: Current changes in tropical
- 29 precipitation. *Environmental Research Letters*, **5**, 025205
- 30 Allen, M. R., and W. J. Ingram, 2002: Constraints on future changes in climate and the hydrologic cycle.
- 31 *Nature*, **419**, 224-232.
- 32 Allen, R. J., and S. C. Sherwood, 2010: Aerosol-cloud semi-direct effect and land-sea temperature contrast in
- 33 a GCM. *Geophysical Research Letters*, **37**, L07702.
- 34 Anderson, T. L., R. J. Charlson, S. E. Schwartz, R. Knutti, O. Boucher, H. Rodhe, and J. Heintzenberg,
- 35 2003: Climate forcing by aerosols - a hazy picture. *Science*, **300**, 1103-1104.
- 36 Anderson, T. L., and Coauthors, 2009: Temporal and Spatial variability of clouds and related aerosol. *Clouds*
- 37 *in the Perturbed Climate System: Their Relationship to Energy Balance, Atmospheric Dynamics, and*
- 38 *Precipitation*, R. J. a. H. Charlson, J., Ed., MIT Press, 127-148.
- 39 Anderson, T. L., and Coauthors, 2005: An "A-Train" strategy for quantifying direct climate forcing by
- 40 anthropogenic aerosols. *Bulletin of the American Meteorological Society*, **86**, 1795-1809.
- 41 Andreae, M. O., and A. Gelencser, 2006: Black carbon or brown carbon? The nature of light-absorbing
- 42 carbonaceous aerosols. *Atmospheric Chemistry and Physics*, **6**, 3131-3148.
- 43 Andreae, M. O., C. D. Jones, and P. M. Cox, 2005: Strong present-day aerosol cooling implies a hot future.
- 44 *Nature*, **435**, 1187-1190.
- 45 Andreae, M. O., D. Rosenfeld, P. Artaxo, A. A. Costa, G. P. Frank, K. M. Longo, and M. A. F. Silva-Dias,
- 46 2004: Smoking rain clouds over the Amazon. *Science*, **303**, 1337-1342.
- 47 Andrews, T., and P. M. Forster, 2008: CO₂ forcing induces semi-direct effects with consequences for climate
- 48 feedback interpretations. *Geophysical Research Letters*, **35**, ARTN L04802.
- 49 Andrews, T., P. M. Forster, O. Boucher, N. Bellouin, and A. Jones, 2010: Precipitation, radiative forcing and
- 50 global temperature change. *Geophysical Research Letters*, **37**, ARTN L14701.
- 51 Andronova, N. G., and M. E. Schlesinger, 2001: Objective estimation of the probability density function for
- 52 climate sensitivity. *Journal of Geophysical Research*, **106**, 22605-22611.
- 53 Ansmann, A., and Coauthors, 2008: Influence of Saharan dust on cloud glaciation in southern Morocco
- 54 during the Saharan Mineral Dust Experiment. *Journal of Geophysical Research*, **113**, D04210.
- 55 Arakawa, A., 1975: Modeling clouds and cloud processes for use in climate models. *The physical basis of*
- 56 *climate and climate modelling*, ICSU/W MO, 181-197.

- 1 ———, 2004: The cumulus parameterization problem: Past, present, and future. *Journal of Climate*, **17**, 2493-
2 2525.
- 3 Archuleta, C. M., P. J. DeMott, and S. M. Kreidenweis, 2005: Ice nucleation by surrogates for atmospheric
4 mineral dust and mineral dust/sulfate particles at cirrus temperatures. *Atmospheric Chemistry and*
5 *Physics*, **5**, 2617-2634.
- 6 Ariya, P. A., J. Sun, N. A. Eltouny, E. D. Hudson, C. T. Hayes, and G. Kos, 2009: Physical and chemical
7 characterization of bioaerosols - Implications for nucleation processes. *International Reviews in*
8 *Physical Chemistry*, **28**, 1-32.
- 9 Arneth, A., R. K. Monson, G. Schurgers, U. Niinemets, and P. I. Palmer, 2008: Why are estimates of global
10 terrestrial isoprene emissions so similar (and why is this not so for monoterpenes)? *Atmospheric*
11 *Chemistry and Physics*, **8**, 4605-4620.
- 12 Arneth, A., P. A. Miller, M. Scholze, T. Hickler, G. Schurgers, B. Smith, and I. C. Prentice, 2007: CO₂
13 inhibition of global terrestrial isoprene emissions: Potential implications for atmospheric chemistry.
14 *Geophysical Research Letters*, **34**, ARTN L18813.
- 15 Arnold, F., 2006: Atmospheric aerosol and cloud condensation nuclei formation: A possible influence of
16 cosmic rays? *Space Science Reviews*, **125**, 169-186.
- 17 Artaxo, P., and Coauthors, 1998: Large-scale aerosol source apportionment in Amazonia. *Journal of*
18 *Geophysical Research*, **103**, 31837-31847.
- 19 Artaxo, P., and Coauthors, 2002: Physical and chemical properties of aerosols in the wet and dry seasons in
20 Rondônia, Amazonia. *Journal of Geophysical Research*, **107**.
- 21 Aw, J., and M. J. Kleeman, 2003: Evaluating the first-order effect of intraannual temperature variability on
22 urban air pollution. *Journal of Geophysical Research*, **108**, ARTN 4365.
- 23 Ayers, G. P., and J. M. Caine, 2007: The CLAW hypothesis: a review of the major developments.
24 *Environmental Chemistry*, **4**, 366-374.
- 25 Baker, M. B., and R. J. Charlson, 1990: Bistability of CCN concentrations and thermodynamics in the cloud-
26 topped boundary-layer. *Nature*, **345**, 142-145.
- 27 Baker, M. B., and T. Peter, 2008: Small-scale cloud processes and climate. *Nature*, **451**, 299-300.
- 28 Bala, G., P. B. Duffy, and K. E. Taylor, 2008: Impact of geoengineering schemes on the global hydrological
29 cycle. *Proc. Natl. Acad. Sci. U S A*, **105**, 7664-7669.
- 30 Bala, G., K. Caldeira, and R. Nemani, 2010: Fast versus slow response in climate change: implications for
31 the global hydrological cycle. *Climate Dynamics*, **35**, 423-434.
- 32 Barahona, D., and A. Nenes, 2009: Parameterizing the competition between homogeneous and
33 heterogeneous freezing in ice cloud formation - polydisperse ice nuclei. *Atmospheric Chemistry and*
34 *Physics*, **9**, 5933-5948.
- 35 Barahona, D., R. Sotiropoulou, and A. Nenes, 2011: Global distribution of cloud droplet number
36 concentration, autoconversion rate, and aerosol indirect effect under diabatic droplet activation.
37 *Journal of Geophysical Research*, **116**, D09203.
- 38 Barker, T., and Coauthors, 2007: *Summary for Policymakers, IPCC Fourth Assessment Report, Working*
39 *Group III*. 36 pp.
- 40 Barmet, P., T. Kuster, A. Muehler, and U. Lohmann, 2009: Weekly cycle in particulate matter versus
41 weekly cycle in precipitation over Switzerland. *J. Geophys. Res.*, **114**, D05206.
- 42 Bauer, S., E. Bierwirth, M. Esselborn, A. Petzold, A. Macke, T. Trautmann, and M. Wendisch, 2011:
43 Airborne spectral radiation measurements to derive solar radiative forcing of Saharan dust mixed with
44 biomass burning smoke particles. *Tellus*, **63B**, 742-750.
- 45 Bauer, S. E., D. Koch, N. Unger, S. M. Metzger, D. T. Shindell, and D. G. Streets, 2007: Nitrate aerosols
46 today and in 2030: a global simulation including aerosols and tropospheric ozone. *Atmospheric*
47 *Chemistry and Physics*, **7**, 5043-5059.
- 48 Bauer, S. E., and Coauthors, 2008: MATRIX (Multiconfiguration Aerosol TRacker of mIXing state): an
49 aerosol microphysical module for global atmospheric models. *Atmospheric Chemistry and Physics*, **8**,
50 6003-6035.
- 51 Bäumer, D., R. Rinke, and B. Vogel, 2008: Weekly periodicities of Aerosol Optical Thickness over Central
52 Europe – evidence of an anthropogenic direct aerosol effect. *Atmos. Chem. Phys.*, **8**, 83-90.
- 53 Baustian, K. J., M. E. Wise, and M. A. Tolbert, 2010: Depositional ice nucleation on solid ammonium
54 sulfate and glutaric acid particles. *Atmospheric Chemistry and Physics*, **10**, 2307-2317.
- 55 Bazilevskaya, G. A., and Coauthors, 2008: Cosmic ray induced ion production in the atmosphere. *Space*
56 *Science Reviews*, **137**, 149-173.

- 1 Bechtold, P., and Coauthors, 2008: Advances in simulating atmospheric variability with the ECMWF model:
2 From synoptic to decadal time-scales. *Quarterly Journal of the Royal Meteorological Society*, **134**,
3 1337-1351.
- 4 Bell, T. L., D. Rosenfeld, K.-M. Kim, J.-M. Yoo, M.-I. Lee, and M. Hahnenberger, 2008: Midweek increase
5 in US summer rain and storm heights suggests air pollution invigorates rainstorms. *Journal of*
6 *Geophysical Research-Atmospheres*, **113**.
- 7 Bellouin, N., O. Boucher, J. Haywood, and M. S. Reddy, 2005: Global estimate of aerosol direct radiative
8 forcing from satellite measurements. *Nature*, **438**, 1138-1141.
- 9 Bellouin, N., A. Jones, J. Haywood, and S. A. Christopher, 2008: Updated estimate of aerosol direct
10 radiative forcing from satellite observations and comparison against the Hadley Centre climate model.
11 *Journal of Geophysical Research*, **113**, D10205.
- 12 Bellouin, N., J. Rae, C. Johnson, J. Haywood, A. Jones, and O. Boucher, 2011: Aerosol forcing in Hadley
13 Centre CMIP5 simulations and the role of nitrate. *Journal of Geophysical Research*, **116**, D20206.
- 14 Bender, F. A.-M., V. Ramanathan, and G. Tselioudis, 2011: Changes in extratropical storm track cloudiness
15 1983–2008: observational support for a poleward shift. *Climate Dynamics*, Published online.
- 16 Benedetti, A., and Coauthors, 2009: Aerosol analysis and forecast in the ECMWF Integrated Forecast
17 System. Part II : Data assimilation. *Journal of Geophysical Research*, **114**, D13205.
- 18 Benedict, J. J., and D. A. Randall, 2009: Structure of the Madden-Julian Oscillation in the
19 Superparameterized CAM. *Journal of the Atmospheric Sciences*, **66** 3277-3296.
- 20 Benson, D. R., J. H. Yu, A. Markovich, and S. H. Lee, 2011: Ternary homogeneous nucleation of H₂SO₄,
21 NH₃, and H₂O under conditions relevant to the lower troposphere. *Atmospheric Chemistry and*
22 *Physics*, **11**, 4755-4766.
- 23 Berg, P., J. O. Haerter, P. Thejll, C. Piani, S. Hagemann, and J. H. Christensen, 2009: Seasonal
24 characteristics of the relationship between daily precipitation intensity and surface temperature.
25 *Journal of Geophysical Research*, **114**, D18102.
- 26 Bergamo, A., A. M. Tafuro, S. Kinne, F. De Tomasi, and M. R. Perrone, 2008: Monthly-averaged
27 anthropogenic aerosol direct radiative forcing over the Mediterranean based on AERONET aerosol
28 properties. *Atmospheric Chemistry and Physics*, **8**, 6995-7014.
- 29 Berndt, T., and Coauthors, 2010: Laboratory study on new particle formation from the reaction OH + SO₂:
30 influence of experimental conditions, H₂O vapour, NH₃ and the amine tert-butylamine on the overall
31 process. *Atmospheric Chemistry and Physics*, **10**, 7101-7116.
- 32 Bertram, A. K., T. Koop, L. T. Molina, and M. J. Molina, 2000: Ice formation in (NH₄)₂SO₄-H₂O particles.
33 *Journal of Physical Chemistry A*, **104**, 584-588.
- 34 Bi, X., and Coauthors, 2011: Mixing state of biomass burning particles by single particle aerosol mass
35 spectrometer in the urban area of PRD, China. *Atmospheric Environment*, **45**, 3447-3453.
- 36 Blossey, P. N., C. S. Bretherton, and M. C. Wyant, 2009: Subtropical low cloud response to a warmer
37 climate in a superparameterized climate model: Part II, Column modeling with a cloud resolving
38 model. *J. Adv. Modeling Earth Syst.*, **1**, 8.
- 39 Bond, T. C., and R. W. Bergstrom, 2006: Light absorption by carbonaceous particles: An investigative
40 review. *Aerosol Science and Technology*, **40**, 27-67.
- 41 Bondo, T., M. B. Enghoff, and H. Svensmark, 2010: Model of optical response of marine aerosols to
42 Forbush decreases. *Atmospheric Chemistry and Physics*, **10**, 2765-2776.
- 43 Bony, S., and J. L. Dufresne, 2005: Marine boundary layer clouds at the heart of tropical cloud feedback
44 uncertainties in climate models. *Geophysical Research Letters*, **32**.
- 45 Bony, S., J. L. Dufresne, H. Le Treut, J. J. Morcrette, and C. Senior, 2004: On dynamic and thermodynamic
46 components of cloud changes. *Climate Dynamics*, **22**, 71-86.
- 47 Bony, S., and Coauthors, 2006: How well do we understand and evaluate climate change feedback
48 processes? *Journal of Climate*, **19**, 3445-3482.
- 49 Bopp, L., O. Boucher, O. Aumont, S. Belviso, J. L. Dufresne, M. Pham, and P. Monfray, 2004: Will marine
50 dimethylsulfide emissions amplify or alleviate global warming? A model study. *Canadian Journal of*
51 *Fisheries and Aquatic Sciences*, **61**, 826-835.
- 52 Boucher, O., 1999: Air traffic may increase cirrus cloudiness. *Nature*, 30-31.
- 53 Boucher, O., and U. Lohmann, 1995: The sulfate-CCN-cloud albedo effect - A sensitivity study with 2
54 general circulation models. *Tellus*, **47B**, 281-300.
- 55 Boucher, O., and M. Pham, 2002: History of sulfate aerosol radiative forcings. *Geophysical Research*
56 *Letters*, ARTN 1308.

- 1 Boucher, O., G. Myhre, and A. Myhre, 2004: Direct influence of irrigation on atmospheric water vapour and
2 climate. *Climate Dynamics*, **22**, 597–603.
- 3 Bourotte, C., A.-P. Curi-Amarante, M.-C. Forti, L. A. A. Pereira, A. L. Braga, and P. A. Lotufo, 2007:
4 Association between ionic composition of fine and coarse aerosol soluble fraction and peak expiratory
5 flow of asthmatic patients in Sao Paulo city (Brazil). *Atmospheric Environment*, **41**, 2036–2048.
- 6 Brenninkmeijer, C. A. M., and Coauthors, 2007: Civil Aircraft for the regular investigation of the
7 atmosphere based on an instrumented container: The new CARIBIC system. *Atmospheric Chemistry
8 and Physics*, **7**, 4953–4976.
- 9 Bretherton, C., and S. Park, 2009: A New Moist Turbulence Parameterization in the Community Atmosphere
10 Model. *Journal of Climate*, 3422–3448.
- 11 Bretherton, C. S., P. N. Blossey, and J. Uchida, 2007: Cloud droplet sedimentation, entrainment efficiency,
12 and subtropical stratocumulus albedo. *Geophysical Research Letters*, **34**, L03813.
- 13 Brioude, J., and Coauthors, 2009: Effect of biomass burning on marine stratocumulus clouds off the
14 California coast. *Atmospheric Chemistry and Physics*, **9**, 8841–8856.
- 15 Brock, C. A., and Coauthors, 2011: Characteristics, sources, and transport of aerosols measured in spring
16 2008 during the aerosol, radiation, and cloud processes affecting Arctic Climate (ARCPAC) Project.
17 *Atmospheric Chemistry and Physics*, **11**, 2423–2453.
- 18 Budyko, M. I., 1974: *Izmeniya Klimata*. Gidrometeoizdat.
- 19 Bundke, U., B. Nillius, R. Jaenicke, T. Wetter, H. Klein, and H. Bingemer, 2008: The fast Ice Nucleus
20 chamber FINCH. *Atmospheric Research*, **90**, 180–186.
- 21 Burkhardt, U., and B. Kärcher, 2009: Process-based simulation of contrail cirrus in a global climate model.
22 *J. Geophys. Res.*, **114**, D16201.
- 23 Burkhardt, U., and B. Kärcher, 2011: Global radiative forcing from contrail cirrus. *Nature Climate Change*,
24 **1**, 54–58.
- 25 Caldeira, K., and L. Wood, 2008: Global and Arctic climate engineering: numerical model studies.
26 *Philosophical Transactions of the Royal Society a-Mathematical Physical and Engineering Sciences*,
27 **366**, 4039–4056.
- 28 Caldwell, P., and C. S. Bretherton, 2009: Response of a subtropical stratocumulus-capped mixed layer to
29 climate and aerosol changes. *Journal of Climate*, **22**, 20–38
- 30 Calogovic, J., C. Albert, F. Arnold, J. Beer, L. Desorgher, and E. O. Flueckiger, 2010: Sudden cosmic ray
31 decreases: No change of global cloud cover. *Geophysical Research Letters*, **37**, L03802.
- 32 Cao, L., G. Bala, and K. Caldeira, 2011: Why is there a short-term increase in global precipitation in
33 response to diminished CO₂ forcing? *Geophys. Res. Lett.*, **38**, L06703.
- 34 Carrico, C. M., M. H. Bergin, A. B. Shrestha, J. E. Dibb, L. Gomes, and J. M. Harris, 2003: The importance
35 of carbon and mineral dust to seasonal aerosol properties in the Nepal Himalaya. *Atmospheric
36 Environment*, **37**, 2811–2824.
- 37 Carslaw, K., O. Boucher, D. Spracklen, G. Mann, J. Rae, S. Woodward, and M. Kulmala, 2010: A review of
38 natural aerosol interactions and feedbacks within the Earth system. *Atmospheric Chemistry and
39 Physics*, **10**, 1701–1737.
- 40 Carslaw, K. S., R. G. Harrison, and J. Kirkby, 2002: Cosmic rays, clouds, and climate. *Science*, **298**, 1732–
41 1737.
- 42 Celis, J. E., J. R. Morales, C. A. Zarorc, and J. C. Inzunza, 2004: A study of the particulate matter PM₁₀
43 composition in the atmosphere of Chillán, Chile. *Chemosphere*, **54**, 541–550.
- 44 Cess, R. D., and Coauthors, 1989: Interpretation of cloud-climate feedbacks as produced by 14 atmospheric
45 general circulation models. *Science*, **245**, 513–516.
- 46 Chae, J. H., and S. C. Sherwood, 2010: Insights into cloud-top height and dynamics from the seasonal cycle
47 of cloud-top heights observed by MISR in the West Pacific region. *Journal of the Atmospheric
48 Sciences*, **67**, 248–261.
- 49 Chakraborty, A., and T. Gupta, 2010: Chemical characterization and source apportionment of submicron
50 (PM₁) aerosol in Kanpur region, India. *Aerosol and Air Quality Research*, **10**, 433–445.
- 51 Chameides, W. L., C. Luo, R. Saylor, D. Streets, Y. Huang, M. Bergin, and F. Giorgi, 2002: Correlation
52 between model-calculated anthropogenic aerosols and satellite-derived cloud optical depths:
53 Indication of indirect effect? *Journal of Geophysical Research*, **107**.
- 54 Chan, Y. C., R. W. Simpson, G. H. Mctainsh, P. D. Vowles, D. D. Cohen, and G. M. Bailey, 1997:
55 Characterisation of chemical species in PM_{2.5} and PM₁₀ aerosols in Brisbane, Australia. *Atmospheric
56 Environment*, **31**, 3773–3785.

- 1 Chand, D., R. Wood, T. L. Anderson, S. K. Satheesh, and R. J. Charlson, 2009: Satellite-derived direct
2 radiative effect of aerosols dependent on cloud cover. *Nature Geoscience*, **2**, 181-184.
- 3 Chang, F., and J. Coakley, 2007: Relationships between marine stratus cloud optical depth and temperature:
4 Inferences from AVHRR observations. *Journal of Climate*, 2022-2036.
- 5 Charlson, R. J., A. S. Ackerman, F. A. M. Bender, T. L. Anderson, and Z. Liu, 2007: On the climate forcing
6 consequences of the albedo continuum between cloudy and clear air. *Tellus*, **59B**, 715-727.
- 7 Charney, J. G., 1979: *Carbon dioxide and climate: A scientific assessment*, 33 pp. pp.
- 8 Chen, L., G. Shi, S. Qin, S. Yang, and P. Zhang, 2011: Direct radiative forcing of anthropogenic aerosols
9 over oceans from satellite observations. *Advances in Atmospheric Sciences*, **28**, 973-984.
- 10 Chen, W. T., Y. H. Lee, P. J. Adams, A. Nenes, and J. H. Seinfeld, 2010: Will black carbon mitigation
11 dampen aerosol indirect forcing? *Geophysical Research Letters*, ARTN L09801.
- 12 Chen, Y., and J. E. Penner, 2005: Uncertainty analysis for estimates of the first indirect aerosol effect.
13 *Atmospheric Chemistry and Physics*, **5**, 2935-2948.
- 14 Chen, Y., Q. Li, R. A. Kahn, J. T. Randerson, and D. J. Diner, 2009: Quantifying aerosol direct radiative
15 effect with Multiangle Imaging Spectroradiometer observations: Top-of-atmosphere albedo change by
16 aerosols based on land surface types. *Journal of Geophysical Research*, **114**, D02109.
- 17 Cheng, T., Y. Peng, J. Feichter, and I. Tegen, 2008: An improvement on the dust emission scheme in the
18 global aerosol-climate model ECHAM5-HAM. *Atmospheric Chemistry and Physics*, **8**, 1105-1117.
- 19 Cheng, Z. L., K. S. Lam, L. Y. Chan, and K. K. Cheng, 2000: Chemical characteristics of aerosols at coastal
20 station in Hong Kong. I. Seasonal variation of major ions, halogens and mineral dusts between 1995
21 and 1996. *Atmospheric Environment*, **34**, 2771-2783.
- 22 Chernoff, D. I., and A. K. Bertram, 2010: Effects of sulfate coatings on the ice nucleation properties of a
23 biological ice nucleus and several types of minerals. *Journal of Geophysical Research*, **115**, D20205.
- 24 Chikira, M., and M. Sugiyama, 2010: A cumulus parameterization with state-dependent entrainment rate.
25 Part I: Description and sensitivity to temperature and humidity profiles. *Journal of the Atmospheric
26 Sciences*, **67**, 2171-2193.
- 27 Choi, Y. S., R. S. Lindzen, C. H. Ho, and J. Kim, 2010: Space observations of cold-cloud phase change.
28 *Proceedings of the National Academy of Sciences of the United States of America*, **107**, 11211-11216.
- 29 Chow, J. C., J. G. Waston, D. H. Lowenthal, P. A. Solomon, K. L. Magliano, S. D. Ziman, and L. W.
30 Richards, 1993: PM₁₀ and PM_{2.5} compositions in California's San Joaquin Valley. *Aerosol Science and
31 Technology*, **18**, 105-128.
- 32 Chuang, C. C., J. E. Penner, K. E. Taylor, A. S. Grossman, and J. J. Walton, 1997: An assessment of the
33 radiative effects of anthropogenic sulfate. *Journal of Geophysical Research-Atmospheres*, **102**, 3761-
34 3778.
- 35 Chuang, C. C., J. E. Penner, J. M. Prospero, K. E. Grant, G. H. Rau, and K. Kawamoto, 2002: Cloud
36 susceptibility and the first aerosol indirect forcing: Sensitivity to black carbon and aerosol
37 concentrations. *Journal of Geophysical Research*, **107**, 4564.
- 38 Church, J. A., and Coauthors, 2011: Revisiting the Earth's sea-level and energy budgets from 1961 to 2008.
39 *Geophysical Research Letters*, **38**, L18601.
- 40 Clarke, A. D., V. N. Kapustin, F. L. Eisele, R. J. Weber, and P. H. McMurry, 1999: Particle production near
41 marine clouds: Sulfuric acid and predictions from classical binary nucleation. *Geophysical Research
42 Letters*, **26**, 2425-2428.
- 43 Clement, A. C., R. Burgman, and J. R. Norris, 2009: Observational and model evidence for positive low-
44 level cloud feedback. *Science*, **325**, 460-464.
- 45 Colman, R. A., and B. J. McAvaney, 2011: On tropospheric adjustment to forcing and climate feedbacks.
46 *Climate Dynamics*, **36**, 1649-1658.
- 47 Comstock, K. K., C. S. Bretherton, and S. E. Yuter, 2005: Mesoscale variability and drizzle in Southeast
48 Pacific stratocumulus. *Journal of the Atmospheric Sciences*, **62**, 3792-3807.
- 49 Connolly, P. J., O. Mohler, P. R. Field, H. Saathoff, R. Burgess, T. Choullarton, and M. Gallagher, 2009:
50 Studies of heterogeneous freezing by three different desert dust samples. *Atmospheric Chemistry and
51 Physics*, **9**, 2805-2824.
- 52 Cozic, J., and Coauthors, 2008: Black carbon enrichment in atmospheric ice particle residuals observed in
53 lower tropospheric mixed phase clouds. *Journal of Geophysical Research*, **113**, D15209.
- 54 Crawford, I., and Coauthors, 2011: Studies of propane flame soot acting as heterogeneous ice nuclei in
55 conjunction with single particle soot photometer measurements. *Atmospheric Chemistry and Physics*,
56 **11**, 9549-9561.

- 1 Cross, E. S., and Coauthors, 2010: Soot Particle Studies—Instrument Inter-Comparison—Project Overview.
2 *Aerosol Science and Technology*, **44**, 592-611.
- 3 Crucifix, M., 2006: Does the Last Glacial Maximum constrain climate sensitivity? *Geophysical Research*
4 *Letters*, **33**, L18701.
- 5 Crutzen, P. J., 2006: Albedo enhancement by stratospheric sulfur injections: A contribution to resolve a
6 policy dilemma? *Climatic Change*, **77**, 211-220.
- 7 Cziczo, D. J., K. D. Froyd, S. J. Gallavardin, O. Moehler, S. Benz, H. Saathoff, and D. M. Murphy, 2009a:
8 Deactivation of ice nuclei due to atmospherically relevant surface coatings. *Environmental Research*
9 *Letters*, **4**.
- 10 Cziczo, D. J., and Coauthors, 2009b: Inadvertent climate modification due to anthropogenic lead. *Nature*
11 *Geoscience*, **2**, 333-336.
- 12 Darmenova, K., I. N. Sokolik, Y. P. Shao, B. Marticorena, and G. Bergametti, 2009: Development of a
13 physically based dust emission module within the Weather Research and Forecasting (WRF) model:
14 Assessment of dust emission parameterizations and input parameters for source regions in Central and
15 East Asia. *Journal of Geophysical Research*, **114**, ARTN D14201.
- 16 Dawson, J. P., P. J. Adams, and S. N. Pandis, 2007: Sensitivity of PM_{2.5} to climate in the Eastern US: a
17 modeling case study. *Atmospheric Chemistry and Physics*, **7**, 4295-4309.
- 18 de Boer, G., H. Morrison, M. D. Shupe, and R. Hildner, 2011: Evidence of liquid dependent ice nucleation in
19 high-latitude stratiform clouds from surface remote sensors. *Geophysical Research Letters*, **38**,
20 L01803.
- 21 de Gouw, J. A., and Coauthors, 2005: Budget of organic carbon in a polluted atmosphere: Results from the
22 New England Air Quality Study in 2002. *Journal of Geophysical Research*, **110**, ARTN D16305.
- 23 de Leeuw, G., and Coauthors, 2011: Production flux of sea spray aerosol. *Reviews of Geophysics*, **49**,
24 RG2001.
- 25 Deboudt, K., P. Flament, M. Choel, A. Gloter, S. Sobanska, and C. Colliex, 2010: Mixing state of aerosols
26 and direct observation of carbonaceous and marine coatings on African dust by individual particle
27 analysis. *Journal of Geophysical Research*, **115**, D24207.
- 28 Decesari, S., and Coauthors, 2010: Chemical composition of PM₁₀ and PM₁ at the high-altitude Himalayan
29 station Nepal Climate Observatory-Pyramid (NCO-P) (5079 m.a.s.l.). *Atmospheric Chemistry and*
30 *Physics*, **10**, 4583-4596.
- 31 DeMott, C. A., C. Stan, D. A. Randall, J. L. Kinter III, and M. Khairoutdinov, 2011: The Asian Monsoon in
32 the super-parameterized CCSM and its relation to tropical wave activity. *Journal of Climate*, **24**,
33 5134-5156.
- 34 DeMott, P. J., 1990: An exploratory study of ice nucleation by soot aerosols. *Journal of Applied*
35 *Meteorology*, **29**, 1072-1079.
- 36 DeMott, P. J., Y. Chen, S. M. Kreidenweis, D. C. Rogers, and D. E. Sherman, 1999: Ice formation by black
37 carbon particles. *Geophysical Research Letters*, **26**, 2429-2432.
- 38 DeMott, P. J., M. D. Petters, A. J. Prenni, C. M. Carrico, S. M. Kreidenweis, J. L. Collett, and H.
39 Moosmuller, 2009: Ice nucleation behavior of biomass combustion particles at cirrus temperatures.
40 *Journal of Geophysical Research*, **114**, D16205.
- 41 DeMott, P. J., and Coauthors, 2003: African dust aerosols as atmospheric ice nuclei. *Geophysical Research*
42 *Letters*, **30**, 1732.
- 43 DeMott, P. J., and Coauthors, 2010: Predicting global atmospheric ice nuclei distributions and their impacts
44 on climate. *Proceedings of the National Academy of Sciences of the United States of America*, **107**,
45 11217-11222.
- 46 Dengel, S., D. Aebly, and J. Grace, 2009: A relationship between galactic cosmic radiation and tree rings.
47 *New Phytologist*, **184**, 545-551.
- 48 Denman, K. L., and Coauthors, 2007: Couplings Between Changes in the Climate System and
49 Biogeochemistry. *Climate Change 2007: The Physical Science Basis. Contribution of Working Group*
50 *I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge
51 University Press.
- 52 Derbyshire, S. H., A. V. Maidens, S. F. Milton, R. A. Stratton, and M. R. Willett, 2011: Adaptive
53 detrainment in a convective parametrization. *Q. J. Royal Met. Society*, **137**, 1856-1871.
- 54 Derevianko, G. J., C. Deutsch, and A. Hall, 2009: On the relationship between ocean DMS and solar
55 radiation. *Geophysical Research Letters*, **36**, L17606.
- 56 Dessler, A. E., 2010: A determination of the cloud feedback from climate variations over the past decade.
57 *Science*, **330**, 1523-1527.

- 1 ———, 2011: Cloud variations and the Earth's energy budget. *Geophysical Research Letters*, **38**, L19701.
- 2 Deuzé, J.-L., and Coauthors, 2001: Remote sensing of aerosols over land surfaces from POLDER-ADEOS-1
3 polarized measurements. *Journal of Geophysical Research*, **106**, 4913-4926.
- 4 Devasthale, A., O. Kruger, and H. Graßl, 2005: Change in cloud-top temperatures over Europe. *IEEE*
5 *Geoscience and Remote Sensing Letters*, **2**, 333-336.
- 6 Di Biagio, C., A. di Sarra, and D. Meloni, 2010: Large atmospheric shortwave radiative forcing by
7 Mediterranean aerosols derived from simultaneous ground-based and spaceborne observations and
8 dependence on the aerosol type and single scattering albedo. *Journal of Geophysical Research*, **115**,
9 ARTN D10209.
- 10 Diehl, K., and S. K. Mitra, 1998: A laboratory study of the effects of a kerosene-burner exhaust on ice
11 nucleation and the evaporation rate of ice crystals. *Atmospheric Environment*, **32**, 3145-3151.
- 12 Diehl, K., and S. Wurzler, 2010: Air parcel model simulations of a convective cloud: Bacteria acting as
13 immersion ice nuclei. *Atmospheric Environment*, **44**, 4622-4628.
- 14 Diehl, K., C. Quick, S. Matthias-Maser, S. K. Mitra, and R. Jaenicke, 2001: The ice nucleating ability of
15 pollen - Part I: Laboratory studies in deposition and condensation freezing modes. *Atmospheric*
16 *Research*, **58**, 75-87.
- 17 Doherty, S. J., S. G. Warren, T. C. Grenfell, A. D. Clarke, and R. E. Brandt, 2010: Light-absorbing
18 impurities in Arctic snow. *Atmospheric Chemistry and Physics*, **10**, 11647-11680.
- 19 Donahue, N. M., S. A. Epstein, S. N. Pandis, and A. L. Robinson, 2011: A two-dimensional volatility basis
20 set: 1. Organic-aerosol mixing thermodynamics. *Atmospheric Chemistry and Physics*, **11**, 3303-3318.
- 21 Donner, L. J., and Coauthors, 2011: The dynamical core, physical parameterizations, and basic simulation
22 characteristics of the atmospheric component AM3 of the GFDL global coupled model CM3. *Journal*
23 *of Climate*, **24**, 3484–3519
- 24 Doughty, C. E., C. B. Field, and A. M. S. McMillan, 2011: Can crop albedo be increased through the
25 modification of leaf trichomes, and could this cool regional climate? *Climatic Change*, **104**, 379-387.
- 26 Dubovik, O., and Coauthors, 2011: Statistically optimized inversion algorithm for enhanced retrieval of
27 aerosol properties from spectral multi-angle polarimetric satellite observations. *Atmospheric*
28 *Measurement Techniques*, **4**, 975-1018.
- 29 Dubovik, O., and Coauthors, 2006: Application of spheroid models to account for aerosol particle
30 nonsphericity in remote sensing of desert dust. *Journal of Geophysical Research*, **111**, D11208.
- 31 Duda, D. P., G. L. Stephens, B. Stevens, and W. R. Cotton, 1996: Effects of aerosol and horizontal
32 inhomogeneity on the broadband albedo of marine stratus: Numerical simulations. *Journal of the*
33 *Atmospheric Sciences*, **53**, 3757-3769.
- 34 Dufresne, J.-L., J. Quaas, O. Boucher, S. Denvil, and L. Fairhead, 2005: Contrasts in the effects on climate
35 of anthropogenic sulfate aerosols between the 20th and the 21st century. *Geophysical Research*
36 *Letters*, **32**, ARTN L21703.
- 37 Dufresne, J. L., and S. Bony, 2008: An assessment of the primary sources of spread of global warming
38 estimates from coupled atmosphere-ocean models. *Journal of Climate*, **21**, 5135-5144.
- 39 Dunion, J. P., and C. S. Velden, 2004: The impact of the Saharan air layer on Atlantic tropical cyclone
40 activity. *Bulletin of the American Meteorological Society*, **85**, 353-365.
- 41 Duplissy, J., and Coauthors, 2008: Cloud forming potential of secondary organic aerosol under near
42 atmospheric conditions. *Geophysical Research Letters*, **35**, L03818.
- 43 Durkee, P. A., K. J. Noone, and R. T. Bluth, 2000: The Monterey Area Ship Track experiment. *Journal of*
44 *the Atmospheric Sciences*, **57**, 2523-2541.
- 45 Dusek, U., and Coauthors, 2006: Size matters more than chemistry for cloud-nucleating ability of aerosol
46 particles. *Science*, **312**, 1375-1378.
- 47 Dymarska, M., B. J. Murray, L. M. Sun, M. L. Eastwood, D. A. Knopf, and A. K. Bertram, 2006: Deposition
48 ice nucleation on soot at temperatures relevant for the lower troposphere. *Journal of Geophysical*
49 *Research*, **111**, D04204.
- 50 Easter, R. C., and Coauthors, 2004: MIRAGE: Model description and evaluation of aerosols and trace gases.
51 *Journal of Geophysical Research*, **109**, ARTN D20210.
- 52 Eastman, R., and S. G. Warren, 2010: Interannual variations of Arctic cloud types in relation to sea ice.
53 *Journal of Climate*, **23**, 4216-4232.
- 54 Eastwood, M. L., S. Cremel, M. Wheeler, B. J. Murray, E. Girard, and A. K. Bertram, 2009: Effects of
55 sulfuric acid and ammonium sulfate coatings on the ice nucleation properties of kaolinite particles.
56 *Geophysical Research Letters*, **36**, L02811.

- 1 Eitzen, Z. A., K. M. Xu, and T. Wong, 2009: Cloud and radiative characteristics of tropical deep convective
2 systems in extended cloud objects from CERES observations. *Journal of Climate*, **22**, 5983-6000.
- 3 Eitzen, Z. A., K.-M. Xu, and T. Wong, 2011: An estimate of low-cloud feedbacks from variations of cloud
4 radiative and physical properties with sea surface temperature on interannual time scales. *Journal of*
5 *Climate*, **24**, 1106-1121.
- 6 Enghoff, M. B., and H. Svensmark, 2008: The role of atmospheric ions in aerosol nucleation - a review.
7 *Atmospheric Chemistry and Physics*, **8**, 4911-4923.
- 8 Engström, A., and A. M. L. Ekman, 2010: Impact of meteorological factors on the correlation between
9 aerosol optical depth and cloud fraction. *Geophys. Res. Lett.*, **37**, L18814.
- 10 Ervens, B., G. Feingold, and S. M. Kreidenweis, 2005: Influence of water-soluble organic carbon on cloud
11 drop number concentration. *Journal of Geophysical Research*, **110**, D18211.
- 12 Ervens, B., G. Feingold, K. Sulia, and J. Harrington, 2011: The impact of microphysical parameters, ice
13 nucleation mode, and habit growth on the ice/liquid partitioning in mixed-phase Arctic clouds. *Journal*
14 *of Geophysical Research*, **116**, D17205.
- 15 Evan, A. T., J. Dunion, J. A. Foley, A. K. Heidinger, and C. S. Velden, 2006: New evidence for a
16 relationship between Atlantic tropical cyclone activity and African dust outbreaks. *Geophysical*
17 *Research Letters*, **33**, L19813.
- 18 Evan, A. T., and Coauthors, 2008: Ocean temperature forcing by aerosols across the Atlantic tropical
19 cyclone development region. *Geochemistry Geophysics Geosystems*, **9**, Q05v04.
- 20 Evans, J. R. G., E. P. J. Stride, M. J. Edirisinghe, D. J. Andrews, and R. R. Simons, 2010: Can oceanic foams
21 limit global warming? *Climate Research*, **42**, 155-160.
- 22 Fan, J., R. Zhang, G. Li, J. Nielsen-Gammon, and Z. Li, 2005: Simulations of fine particulate matter (PM_{2.5})
23 in Houston, Texas. *Journal of Geophysical Research*, **110**, D16203.
- 24 Fan, J., and Coauthors, 2009: Dominant role by vertical wind shear in regulating aerosol effects on deep
25 convective clouds. *Journal of Geophysical Research-Atmospheres*, **114**.
- 26 Farina, S. C., P. J. Adams, and S. N. Pandis, 2010: Modeling global secondary organic aerosol formation and
27 processing with the volatility basis set: Implications for anthropogenic secondary organic aerosol.
28 *Journal of Geophysical Research*, **115**, ARTN D09202.
- 29 Farrar, P. D., 2000: Are cosmic rays influencing oceanic cloud coverage – or is it only El Niño? *Climatic*
30 *Change*, **47**, 7-15.
- 31 Favez, O., H. Cachier, J. Sciarea, S. C. Alfaro, T. M. El-Araby, M. A. Harhash, and Magdy M. Abdelwahab,
32 2008: Seasonality of major aerosol species and their transformations in Cairo megacity. *Atmospheric*
33 *Environment*, **42**, 1503–1516.
- 34 Feichter, J., U. Lohmann, and I. Schult, 1997: The atmospheric sulfur cycle in ECHAM-4 and its impact on
35 the shortwave radiation. *Climate Dynamics*, **13**, 235-246.
- 36 Feingold, G., and H. Siebert, 2009: Cloud-aerosol interactions from the micro to the cloud scale. *Clouds in*
37 *the Perturbed Climate System: Their Relationship to Energy Balance, Atmospheric Dynamics, and*
38 *Precipitation*, J. Heintzenberg, and R. J. Charlson, Eds., MIT Press, 319-338.
- 39 Feingold, G., H. L. Jiang, and J. Y. Harrington, 2005: On smoke suppression of clouds in Amazonia.
40 *Geophysical Research Letters*, **32**, ARTN L02804.
- 41 Feingold, G., R. Boers, B. Stevens, and W. R. Cotton, 1997: A modeling study of the effect of drizzle on
42 cloud optical depth and susceptibility. *Journal of Geophysical Research*, **102**, 13527-13534.
- 43 Feldpausch, P., M. Fiebig, L. Fritzsche, and A. Petzold, 2006: Measurement of ultrafine aerosol size
44 distributions by a combination of diffusion screen separators and condensation particle counters.
45 *Journal of Aerosol Science*, **37**, 577-597.
- 46 Feng, J., 2008: A size-resolved model and a four-mode parameterization of dry deposition of atmospheric
47 aerosols. *Journal of Geophysical Research*, **113**, D12201.
- 48 Field, P. R., and Coauthors, 2006: Some ice nucleation characteristics of Asian and Saharan desert dust.
49 *Atmospheric Chemistry and Physics*, **6**, 2991-3006.
- 50 Flanner, M. G., C. S. Zender, J. T. Randerson, and P. J. Rasch, 2007: Present-day climate forcing and
51 response from black carbon in snow. *Journal of Geophysical Research*, **112**, D11202.
- 52 Flanner, M. G., C. S. Zender, P. G. Hess, N. M. Mahowald, T. H. Painter, V. Ramanathan, and P. J. Rasch,
53 2009: Springtime warming and reduced snow cover from carbonaceous particles. *Atmospheric*
54 *Chemistry and Physics*, **9**, 2481-2497.
- 55 Fletcher, N. H., 1962: Surface structure of water and ice. *Philosophical Magazine*, **7**, 255-269.
- 56 Flowers, B. A., M. K. Dubey, C. Mazzoleni, E. A. Stone, J. J. Schauer, S. W. Kim, and S. C. Yoon, 2010:
57 Optical-chemical-microphysical relationships and closure studies for mixed carbonaceous aerosols

- 1 observed at Jeju Island; 3-laser photoacoustic spectrometer, particle sizing, and filter analysis.
2 *Atmospheric Chemistry and Physics*, **10**, 10387-10398.
- 3 Foltz, G. R., and M. J. McPhaden, 2008: Impact of Saharan dust on tropical North Atlantic SST. *Journal of*
4 *Climate*, **21**, 5048-5060.
- 5 Forest, C. E., P. H. Stone, and A. P. Sokolov, 2006: Estimated PDFs of climate system properties including
6 natural and anthropogenic forcings. *Geophysical Research Letters*, **33**, L01705.
- 7 Forest, C. E., P. H. Stone, A. P. Sokolov, M. R. Allen, and M. D. Webster, 2002: Quantifying uncertainties
8 in climate system properties with the use of recent climate observations. *Science*, **295**, 113-117.
- 9 Fornea, A. P., S. D. Brooks, J. B. Dooley, and A. Saha, 2009: Heterogeneous freezing of ice on atmospheric
10 aerosols containing ash, soot, and soil. *Journal of Geophysical Research-Atmospheres*, **114**.
- 11 Forster, P., and Coauthors, 2007: Changes in Atmospheric Constituents and in Radiative Forcing. *Climate*
12 *Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment*
13 *Report of the Intergovernmental Panel on Climate Change*, Cambridge University Press.
- 14 Forster, P. M. D., and J. M. Gregory, 2006: The climate sensitivity and its components diagnosed from Earth
15 Radiation Budget data. *Journal of Climate*, **19**, 39-52.
- 16 Fountoukis, C., and Coauthors, 2007: Aerosol-cloud drop concentration closure for clouds sampled during
17 the International Consortium for Atmospheric Research on Transport and Transformation 2004
18 campaign. *Journal of Geophysical Research*, **112**, D10S30.
- 19 Fowler, L. D., and D. A. Randall, 2002: Interactions between cloud microphysics and cumulus convection in
20 a general circulation model. *Journal of the Atmospheric Sciences*, **59**, 3074-3098.
- 21 Freney, E. J., K. Adachi, and P. R. Buseck, 2010: Internally mixed atmospheric aerosol particles:
22 Hygroscopic growth and light scattering. *Journal of Geophysical Research*, **115**, ARTN D19210.
- 23 Fridlind, A. M., and Coauthors, 2007: Ice properties of single-layer stratocumulus during the Mixed-Phase
24 Arctic Cloud Experiment: 2. Model results. *Journal of Geophysical Research*, **112**, D24202.
- 25 Friedman, B., G. Kulkarni, J. Beranek, A. Zelenyuk, J. A. Thornton, and D. J. Cziczo, 2011: Ice nucleation
26 and droplet formation by bare and coated soot particles. *Journal of Geophysical Research-*
27 *Atmospheres*, **116**, D17203.
- 28 Frömming, C., M. Ponater, U. Burkhardt, A. Stenke, S. Pechtl, and R. Sausen, 2011: Sensitivity of contrail
29 coverage and contrail radiative forcing to selected key parameters. *Atmospheric Environment*, **45**,
30 1483-1490.
- 31 Fuentes, E., H. Coe, D. Green, and G. McFiggans, 2011: On the impacts of phytoplankton-derived organic
32 matter on the properties of the primary marine aerosol - Part 2: Composition, hygroscopicity and cloud
33 condensation activity. *Atmospheric Chemistry and Physics*, **11**, 2585-2602.
- 34 Fuzzi, S., and Coauthors, 2007: Overview of the inorganic and organic composition of size-segregated
35 aerosol in Rondonia, Brazil, from the biomass-burning period to the onset of the wet season. *Journal*
36 *of Geophysical Research*, **112**, D01201.
- 37 Gagen, M., and Coauthors, 2011: Cloud response to summer temperatures in Fennoscandia over the last
38 thousand years. *Geophysical Research Letters*, **38**, 5.
- 39 Gao, R. S., and Coauthors, 2007: A novel method for estimating light-scattering properties of soot aerosols
40 using a modified single-particle soot photometer. *Aerosol Science and Technology*, **41**, 125-135.
- 41 Garcia, O. E., F. J. Exposito, J. P. Diaz, and A. M. Diaz, 2011: Radiative forcing under mixed aerosol
42 conditions. *Journal of Geophysical Research*, **116**, ARTN D01201.
- 43 Gasso, S., 2008: Satellite observations of the impact of weak volcanic activity on marine clouds. *Journal of*
44 *Geophysical Research*, **113**, D14s19.
- 45 Geogdzhayev, I. V., M. I. Mishchenko, W. B. Rossow, B. Cairns, and A. A. Lacis, 2002: Global two-channel
46 AVHRR retrievals of aerosol properties over the ocean for the period of NOAA-9 observations and
47 preliminary retrievals using NOAA-7 and NOAA-11 data. *Journal of the Atmospheric Sciences*, **59**,
48 262-278.
- 49 George, R. C., and R. Wood, 2010: Subseasonal variability of low cloud radiative properties over the
50 southeast Pacific Ocean. *Atmospheric Chemistry and Physics*, **10**, 4047-4063.
- 51 Gerasopoulos, E., and Coauthors, 2007: Size-segregated mass distributions of aerosols over Eastern
52 Mediterranean: seasonal variability and comparison with AERONET columnar size-distributions.
53 *Atmospheric Chemistry and Physics*, **7**, 2551-2561.
- 54 Gettelman, A., J. T. Fasullo, and J. E. Kay, 2011a: Constraining simulated climate sensitivity with cloud
55 microphysics **submitted**.
- 56 Gettelman, A., J. E. Kay, and K. M. Shell, 2011b: The evolution of climate sensitivity and climate feedbacks
57 in the Community Atmosphere Model. *Journal of Climate*, Submitted.

- 1 Gettelman, A., and Coauthors, 2010: Global simulations of ice nucleation and ice supersaturation with an
2 improved cloud scheme in the Community Atmosphere Model. *Journal of Geophysical Research*, **115**,
3 ARTN D18216.
- 4 Ghan, S., R. Easter, J. Hudson, and F. M. Bréon, 2001: Evaluation of aerosol indirect radiative forcing in
5 MIRAGE. *Journal of Geophysical Research*, **106**, 5317-5334.
- 6 Ghan, S. J., X. Liu, R. C. Easter, R. Zaveri, P. J. Rasch, and J.-H. Yoon, 2011a: Toward a minimal
7 representation of aerosol direct, semi-direct and indirect effects: Comparative decomposition. *Journal*
8 *of Climate*, subm. to.
- 9 Ghan, S. J., H. Abdul-Razzak, A. Nenes, Y. Ming, X. Liu, and M. Ovchinnikov, 2011b: Droplet nucleation:
10 Physically-based parameterizations and comparative evaluation. *Journal of Advances in Modeling*
11 *Earth Systems*, **3**, M10001.
- 12 Gioda, A., B. S. Amaral, I. L. GoncalvesMonteiro, and T. D. Saint'Pierre, 2011: Chemical composition,
13 sources, solubility, and transport of aerosol trace elements in a tropical region. *Journal of*
14 *Environmental Monitoring*, **13**, 2134-2142.
- 15 Girard, E., J.-P. Blanchet, and Y. Dubois, 2004: Effects of arctic sulphuric acid aerosols on wintertime low-
16 level atmospheric ice crystals, humidity and temperature at Alert, Nunavut. *Atmospheric Research*, **73**,
17 131-148.
- 18 Givati, A., and D. Rosenfeld, 2004: Quantifying precipitation suppression due to air pollution. *Journal of*
19 *Applied Meteorology*, **43**, 1038-1056.
- 20 Good, N., and Coauthors, 2010: Consistency between parameterisations of aerosol hygroscopicity and CCN
21 activity during the RHaMBLe discovery cruise. *Atmospheric Chemistry and Physics*, **10**, 3189-3203.
- 22 Gorbunov, B., A. Baklanov, N. Kakutkina, H. L. Windsor, and R. Toumi, 2001: Ice nucleation on soot
23 particles. *Journal of Aerosol Science*, **32**, 199-215.
- 24 Gordon, N. D., and J. R. Norris, 2010: Cluster analysis of midlatitude oceanic cloud regimes: mean
25 properties and temperature sensitivity. *Atmospheric Chemistry and Physics*, **10**, 6435-6459.
- 26 Graber, E. R., and Y. Rudich, 2006: Atmospheric HULIS: How humic-like are they? A comprehensive and
27 critical review. *Atmospheric Chemistry and Physics*, **6**, 729-753.
- 28 Grabowski, W. W., 2001: Coupling cloud processes with the large-scale dynamics using the Cloud-
29 Resolving Convection Parameterization (CRCP). *Journal of the Atmospheric Sciences*, **58**, 978-997.
- 30 Grabowski, W. W., and P. K. Smolarkiewicz, 1999: CRCP: a Cloud Resolving Convection Parameterization
31 for modeling the tropical convecting atmosphere. *Physica D*, **133**, 171-178.
- 32 Granier, C., and Coauthors, 2011: Evolution of anthropogenic and biomass burning emissions of air
33 pollutants at global and regional scales during the 1980-2010 period. *Climatic Change*, **109**, 163-190.
- 34 Gregory, J., and M. Webb, 2008: Tropospheric adjustment induces a cloud component in CO2 forcing.
35 *Journal of Climate*, 58-71.
- 36 Gregory, J. M., R. J. Stouffer, S. C. B. Raper, P. A. Stott, and N. A. Rayner, 2002: An observationally based
37 estimate of the climate sensitivity. *Journal of Climate*, **15**, 3117-3121.
- 38 Grote, R., and U. Niinemets, 2008: Modeling volatile isoprenoid emissions - a story with split ends. *Plant*
39 *Biology*, **10**, 8-28.
- 40 Guenther, A., T. Karl, P. Harley, C. Wiedinmyer, P. I. Palmer, and C. Geron, 2006: Estimates of global
41 terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from
42 Nature). *Atmospheric Chemistry and Physics*, **6**, 3181-3210.
- 43 Gullu, H. G., I. Ölmez, and G. Tuncel, 2000: Temporal variability of atmospheric trace element
44 concentrations over the eastern Mediterranean Sea. *Spectrochimica Acta*, **B55**, 1135-1150.
- 45 Haerter, J. O., E. Roeckner, L. Tomassini, and J. S. von Storch, 2009: Parametric uncertainty effects on
46 aerosol radiative forcing. *Geophysical Research Letters*, **36**, L15707.
- 47 Hagler, G. S. W., and Coauthors, 2006: Source areas and chemical composition of fine particulate matter in
48 the Pearl River Delta region of China. *Atmospheric Environment*, **40**, 3802-3815.
- 49 Halfon, N., Z. Levin, and P. Alpert, 2009: Temporal rainfall fluctuations in Israel and their possible link to
50 urban and air pollution effects. *Environmental Research Letters*, **4**, 025001.
- 51 Hallquist, M., and Coauthors, 2009: The formation, properties and impact of secondary organic aerosol:
52 current and emerging issues. *Atmospheric Chemistry and Physics*, **9**, 5155-5236.
- 53 Hamwey, R. M., 2007: Active amplification of the terrestrial albedo to mitigate climate change: An
54 exploratory study. *Mitigation and Adaptation Strategies for Global Change*, **12**, 419-439.
- 55 Han, Y.-J., T.-S. Kim, and H. Kim, 2008: Ionic constituents and source analysis of PM_{2.5} in three Korean
56 cities. *Atmospheric Environment*, **42**, 4735-4746.

- 1 Hand, V. L., G. Capes, D. J. Vaughan, P. Formenti, J. M. Haywood, and H. Coe, 2010: Evidence of internal
2 mixing of African dust and biomass burning particles by individual particle analysis using electron
3 beam techniques. *Journal of Geophysical Research-Atmospheres*, **115**, D13301.
- 4 Hansen, J., and L. Nazarenko, 2004: Soot climate forcing via snow and ice albedos. *Proceedings of the*
5 *National Academy of Sciences of the United States of America*, **101**, 423-428.
- 6 Hansen, J., M. Sato, P. Kharecha, and K. von Schuckmann, 2011: Earth's energy imbalance and implications.
7 *Atmospheric Chemistry and Physics Discussion*, **11**, 27031-27105.
- 8 Hansen, J., and Coauthors, 2005: Efficacy of climate forcings. *Journal of Geophysical Research*, **110**, ARTN
9 D18104.
- 10 Harrington, J. Y., D. Lamb, and R. Carver, 2009: Parameterization of surface kinetic effects for bulk
11 microphysical models: Influences on simulated cirrus dynamics and structure. *Journal of Geophysical*
12 *Research*, **114**, D06212.
- 13 Harrison, R. G., 2008: Discrimination between cosmic ray and solar irradiance effects on clouds, and
14 evidence for geophysical modulation of cloud thickness. *Proceedings of the Royal Society A*, **464**,
15 2575-2590.
- 16 Harrison, R. G., and D. B. Stephenson, 2006: Empirical evidence for a nonlinear effect of galactic cosmic
17 rays on clouds. *Proceedings of the Royal Society A*, **462**, 1221-1233.
- 18 Harrison, R. G., and M. H. P. Ambaum, 2010: Observing Forbush decreases in cloud at Shetland. *Journal of*
19 *Atmospheric and Solar-Terrestrial Physics*, **72**, 1408-1414.
- 20 Hartmann, D. L., and K. Larson, 2002: An important constraint on tropical cloud - climate feedback.
21 *Geophysical Research Letters*, **29**, 1951.
- 22 Harvey, L. D. D., and R. K. Kaufmann, 2002: Simultaneously constraining climate sensitivity and aerosol
23 radiative forcing. *Journal of Climate*, **15**, 2837-2861.
- 24 Haywood, J., and O. Boucher, 2000: Estimates of the direct and indirect radiative forcing due to tropospheric
25 aerosols: A review. *Reviews of Geophysics*, **38**, 513-543.
- 26 Haywood, J., and M. Schulz, 2007: Causes of the reduction in uncertainty in the anthropogenic radiative
27 forcing of climate between IPCC (2001) and IPCC (2007). *Geophysical Research Letters*, **34**, ARTN
28 L20701.
- 29 Haywood, J. M., and Coauthors, 2009: A case study of the radiative forcing of persistent contrails evolving
30 into contrail-induced cirrus. *Journal of Geophysical Research*, **114**, D24201.
- 31 Haywood, J. M., and Coauthors, 2011: Motivation, rationale and key results from the GERBILS Saharan
32 dust measurement campaign. *Quarterly Journal of the Royal Meteorological Society*, **137**, 1106-1116.
- 33 Heald, C. L., D. A. Ridley, S. M. Kreidenweis, and E. E. Drury, 2010: Satellite observations cap the
34 atmospheric organic aerosol budget. *Geophysical Research Letters*, **37**, L24808.
- 35 Heald, C. L., and Coauthors, 2005: A large organic aerosol source in the free troposphere missing from
36 current models. *Geophysical Research Letters*, ARTN L18809.
- 37 Heald, C. L., and Coauthors, 2008: Predicted change in global secondary organic aerosol concentrations in
38 response to future climate, emissions, and land use change. *Journal of Geophysical Research*, **113**,
39 ARTN D05211.
- 40 Heckendorn, P., and Coauthors, 2009: The impact of geoengineering aerosols on stratospheric temperature
41 and ozone. *Environmental Research Letters*, **4**, ARTN 045108.
- 42 Hegerl, G. C., T. J. Crowley, M. Allen, W. T. Hyde, H. N. Pollack, J. Smerdon, and E. Zorita, 2007:
43 Detection of human influence on a new, validated 1500-year temperature reconstruction. *Journal of*
44 *Climate*, **20**, 650-666.
- 45 Hegg, D. A., L. F. Radke, and P. V. Hobbs, 1990: Particle production associated with marine clouds. *Journal*
46 *of Geophysical Research*, **95**, 13917-13926.
- 47 Hegg, D. A., S. G. Warren, T. C. Grenfell, S. J. Doherty, and A. D. Clarke, 2010: Sources of light-absorbing
48 aerosol in Arctic snow and their seasonal variation. *Atmospheric Chemistry and Physics*, **10**, 10923-
49 10938.
- 50 Heintzenberg, J., D. C. Covert, and R. Van Dingenen, 2000: Size distribution and chemical composition of
51 marine aerosols: a compilation and review. *Tellus*, **52**, 1104-1122.
- 52 Held, I. M., and B. J. Soden, 2006: Robust responses of the hydrological cycle to global warming. *Journal of*
53 *Climate*, **19**, 5686-5699.
- 54 Hendricks Franssen, H. J., T. Kuster, P. Barmet, and U. Lohmann, 2009: Comment on "Winter 'weekend
55 effect' in southern Europe and its connection with periodicities in atmospheric dynamics" by A.
56 Sanchez-Lorenzo et al., *Geophys. Res. Lett.*, **36**, L13706.

- 1 Hendricks, J., B. Karcher, U. Lohmann, and M. Ponater, 2005: Do aircraft black carbon emissions affect
2 cirrus clouds on the global scale? *Geophysical Research Letters*, **32**, L12814.
- 3 Heymsfield, A., D. Baumgardner, P. DeMott, P. Forster, K. Gierens, and B. Karcher, 2010: Contrail
4 microphysics. *Bulletin of the American Meteorological Society*, 465-+.
- 5 Heymsfield, A. J., and L. M. Miloshevich, 1995: Relative humidity and temperature influences on cirrus
6 formation and evolution: Observations from wave clouds and FIRE II. *Journal of the Atmospheric
7 Sciences*, **52**, 4302-4326.
- 8 Heymsfield, A. J., and Coauthors, 1998: Cloud properties leading to highly reflective tropical cirrus:
9 Interpretations from CEPEX, TOGA COARE, and Kwajalein, Marshall Islands. *Journal of
10 Geophysical Research-Atmospheres*, **103**, 8805-8812.
- 11 Hill, A. A., and S. Dobbie, 2008: The impact of aerosols on non-precipitating marine stratocumulus. II: The
12 semi-direct effect. *Quarterly Journal of the Royal Meteorological Society*, **134**, 1155-1165.
- 13 Hirsikko, A., and Coauthors, 2011: Atmospheric ions and nucleation: a review of observations. *Atmospheric
14 Chemistry and Physics*, **11**, 767-798.
- 15 Hjellbrekke, A. G., 2001: Data report 1999, Acidifying and eutrophying compounds, Part 1, Annual
16 summaries, 95 pp. pp.
- 17 Holben, B. N., and Coauthors, 1998: AERONET - A federated instrument network and data archive for
18 aerosol characterization. *Remote Sensing of Environment*, **66**, 1-16.
- 19 Holben, B. N., and Coauthors, 2001: An emerging ground-based aerosol climatology: Aerosol optical depth
20 from AERONET. *Journal of Geophysical Research*, **106**, 12067-12097.
- 21 Holloway, C. E., and J. D. Neelin, 2009: Moisture Vertical Structure, Column Water Vapor, and Tropical
22 Deep Convection. *Journal of the Atmospheric Sciences*, **66**, 1665-1683.
- 23 Hoose, C., J. E. Kristjansson, and S. M. Burrows, 2010a: How important is biological ice nucleation in
24 clouds on a global scale? *Environmental Research Letters*, **5**, 024009.
- 25 Hoose, C., U. Lohmann, R. Erdin, and I. Tegen, 2008: The global influence of dust mineralogical
26 composition on heterogeneous ice nucleation in mixed-phase clouds. *Environmental Research Letters*,
27 **3**, 025003.
- 28 Hoose, C., J. E. Kristjansson, J. P. Chen, and A. Hazra, 2010b: A classical-theory-based parameterization of
29 heterogeneous ice nucleation by mineral dust, soot, and biological particles in a global climate model.
30 *Journal of the Atmospheric Sciences*, **67**, 2483-2503.
- 31 Hoose, C., J. E. Kristjansson, T. Iversen, A. Kirkevåg, O. Seland, and A. Gettelman, 2009: Constraining
32 cloud droplet number concentration in GCMs suppresses the aerosol indirect effect. *Geophysical
33 Research Letters*, **36**, L12807.
- 34 Houze, R. A., Jr., 1993: *Climate Dynamics*. Academic Press.
- 35 Hsieh, W. C., A. Nenes, R. C. Flagan, J. H. Seinfeld, G. Buzorius, and H. Jonsson, 2009: Parameterization of
36 cloud droplet size distributions: Comparison with parcel models and observations. *J. Geophys. Res.*,
37 **114**, D11205.
- 38 Hu, M., L. Y. He, Y. H. Zhang, M. Wang, Y. Pyo Kim, and K. C. Moon, 2002: Seasonal variation of ionic
39 species in fine particles at Qingdao. *Atmospheric Environment*, **36**, 5853-5859.
- 40 Huber, M., and R. Knutti, 2011: Probabilistic climate projections with an intermediate complexity model.
41 Part II: Application to observational datasets. *Climate Dynamics*, subm. to.
- 42 Huebert, B. J., and Coauthors, 2010: Linearity of DMS transfer coefficient with both friction velocity and
43 wind speed in the moderate wind speed range. *Geophysical Research Letters*, ARTN L01605.
- 44 Hueglin, C., R. Gehrig, U. Baltensperger, M. Gysel, C. Monn, and H. Vonmont, 2005: Chemical
45 characterisation of PM_{2.5}, PM₁₀ and coarse particles at urban, near-city and rural sites in Switzerland.
46 *Atmospheric Environment*, **39**, 637-651.
- 47 Huffman, G. J., and Coauthors, 2007: The TRMM multisatellite precipitation analysis (TMPA): Quasi-
48 global, multiyear, combined-sensor precipitation estimates at fine scales. *Journal of
49 Hydrometeorology*, **8**, 38-55.
- 50 Huneus, N., F. Chevallier, and O. Boucher, 2011a: Estimating aerosol emissions by assimilating observed
51 aerosol optical depth in a global aerosol model. *Journal of Geophysical Research*, submitted.
- 52 Huneus, N., and Coauthors, 2011b: Global dust model intercomparison in AeroCom phase I, **11**, 7781-
53 7816.
- 54 Iannone, R., D. I. Chernoff, A. Pringle, S. T. Martin, and A. K. Bertram, 2011: The ice nucleation ability of
55 one of the most abundant types of fungal spores found in the atmosphere. *Atmospheric Chemistry and
56 Physics*, **11**, 1191-1201.

- 1 Iga, S., H. Tomita, Y. Tsushima, and M. Satoh, 2010: Sensitivity of Hadley Circulation to physical
2 parameters and resolution through changing upper-tropospheric ice clouds using a global cloud-system
3 resolving model. *Journal of Climate*, **24**, 2666-2679.
- 4 Inoue, T., M. Satoh, Y. Hagihara, H. Miura, and J. Schmetz, 2010: Comparison of high-level clouds
5 represented in a global cloud system-resolving model with CALIPSO/CloudSat and geostationary
6 satellite observations. *Journal of Geophysical Research*, **115**, D00H22.
- 7 IPCC, 2001: *Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third*
8 *Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press,
9 881 pp.
- 10 ———, 2007: *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the*
11 *Fourth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC)*. Cambridge
12 University Press, 996 pp.
- 13 Irvine, P. J., A. Ridgwell, and D. J. Lunt, 2010: Assessing the regional disparities in geoengineering impacts.
14 *Geophysical Research Letters*, **37**, L18702.
- 15 Irvine, P. J., A. Ridgwell, and D. J. Lunt, 2011: Climatic effects of surface albedo geoengineering. *Journal of*
16 *Geophysical Research*, **in press**.
- 17 Irwin, M., N. Good, J. Crosier, T. W. Chouarton, and G. McFiggans, 2010: Reconciliation of measurements
18 of hygroscopic growth and critical supersaturation of aerosol particles in central Germany.
19 *Atmospheric Chemistry and Physics*, **10**, 11737-11752.
- 20 Ito, K., N. Xue, and G. Thurston, 2004: Spatial variation of PM_{2.5} chemical species and source-apportioned
21 mass concentrations in New York City. *Atmospheric Environment*, **38**, 5269-5282.
- 22 Jacob, D. J., and Coauthors, 2003: Transport and Chemical Evolution over the Pacific (TRACE-P) aircraft
23 mission: Design, execution, and first results. *Journal of Geophysical Research*, **108**, 1-19.
- 24 Jacob, D. J., and Coauthors, 2010: The Arctic Research of the Composition of the Troposphere from Aircraft
25 and Satellites (ARCTAS) mission: design, execution, and first results. *Atmospheric Chemistry and*
26 *Physics*, **10**, 5191-5212.
- 27 Jacobson, M. Z., 2001: Strong radiative heating due to the mixing state of black carbon in atmospheric
28 aerosols. *Nature*, **409**, 695-697.
- 29 ———, 2004: Climate response of fossil fuel and biofuel soot, accounting for soot's feedback to snow and sea
30 ice albedo and emissivity. *Journal of Geophysical Research*, **109**, D21201.
- 31 ———, 2006: Effects of externally-through-internally-mixed soot inclusions within clouds and precipitation on
32 global climate. *Journal of Physical Chemistry A*, **110**, 6860-6873.
- 33 Jaegle, L., P. K. Quinn, T. S. Bates, B. Alexander, and J. T. Lin, 2011: Global distribution of sea salt
34 aerosols: new constraints from in situ and remote sensing observations. *Atmospheric Chemistry and*
35 *Physics*, **11**, 3137-3157.
- 36 Jakob, C., and C. Schumacher, 2008: Precipitation and latent heating characteristics of the major tropical
37 western Pacific cloud regimes. *Journal of Climate*, **21**, 4348-4364.
- 38 Jakob, C., G. Tselioudis, and T. Hume, 2005: The radiative, cloud, and thermodynamic properties of the
39 major tropical western Pacific cloud regimes. *Journal of Climate*, **18**, 1203-1215.
- 40 Jenkins, G. S., A. S. Pratt, and A. Heymsfield, 2008: Possible linkages between Saharan dust and tropical
41 cyclone rain band invigoration in the eastern Atlantic during NAMMA-06. *Geophysical Research*
42 *Letters*, **35**, 7.
- 43 Jeong, M. J., and Z. Q. Li, 2005: Quality, compatibility, and synergy analyses of global aerosol products
44 derived from the Advanced Very High Resolution Radiometer and Total Ozone Mapping
45 Spectrometer. *Journal of Geophysical Research*, **110**, ARTN D10S08.
- 46 Jethva, H., S. K. Satheesh, J. Srinivasan, and K. K. Moorthy, 2009: How good is the assumption about
47 visible surface reflectance in MODIS aerosol retrieval over land? A comparison with aircraft
48 measurements over an urban site in India. *IEEE Transactions on Geoscience and Remote Sensing*, **47**,
49 1990-1998.
- 50 Jiang, H. L., H. W. Xue, A. Teller, G. Feingold, and Z. Levin, 2006: Aerosol effects on the lifetime of
51 shallow cumulus. *Geophysical Research Letters*, **33**, L14806.
- 52 Jiang, J. H., H. Su, M. Schoeberl, S. T. Massie, P. Colarco, S. Platnick, and N. J. Livesey, 2008: Clean and
53 polluted clouds: relationships among pollution, ice cloud and precipitation in South America.
54 *Geophys. Res. Lett.*, **35**, L14804.
- 55 Jickells, T. D., and Coauthors, 2005: Global iron connections between desert dust, ocean biogeochemistry,
56 and climate. *Science*, **308**, 67-71.

- 1 Jimenez, J. L., and Coauthors, 2009: Evolution of organic aerosols in the atmosphere. *Science*, **326**, 1525-
2 1529.
- 3 Jirak, I. L., and W. R. Cotton, 2006: Effect of air pollution on precipitation along the front range of the
4 Rocky Mountains. *Journal of Applied Meteorology and Climatology*, **45**, 236-245.
- 5 Johns, T. C., and Coauthors, 2006: The new Hadley Centre Climate Model (HadGEM1): Evaluation of
6 coupled simulations. *Journal of Climate*, **19**, 1327-1353.
- 7 Johnson, B. T., 2005: The semidirect aerosol effect: Comparison of a single-column model with large eddy
8 simulation for marine stratocumulus. *Journal of Climate*, **18**, 119-130.
- 9 Johnson, B. T., K. P. Shine, and P. M. Forster, 2004: The semi-direct aerosol effect: Impact of absorbing
10 aerosols on marine stratocumulus. *Quarterly Journal of the Royal Meteorological Society*, **130**, 1407-
11 1422.
- 12 Johnson, N. C., and S. P. Xie, 2010: Changes in the sea surface temperature threshold for tropical
13 convection. *Nature Geoscience*, **3**, 842-845.
- 14 Jones, A., D. L. Roberts, and A. Slingo, 1994: A climate model study of indirect radiative forcing by
15 anthropogenic sulfate aerosols. *Nature*, **370**, 450-453.
- 16 Jones, A., J. M. Haywood, and O. Boucher, 2007: Aerosol forcing, climate response and climate sensitivity
17 in the Hadley Centre climate model. *Journal of Geophysical Research*, **112**, ARTN D20211.
- 18 Jones, A., J. Haywood, and O. Boucher, 2009: Climate impacts of geoengineering marine stratocumulus
19 clouds. *Journal of Geophysical Research*, **114**, ARTN D10106.
- 20 Jones, A., D. L. Roberts, M. J. Woodage, and C. E. Johnson, 2001: Indirect sulphate aerosol forcing in a
21 climate model with an interactive sulphur cycle. *Journal of Geophysical Research*, **106**, 20293-20310.
- 22 Jones, A., J. Haywood, O. Boucher, B. Kravitz, and A. Robock, 2010a: Geoengineering by stratospheric SO₂
23 injection: results from the Met Office HadGEM(2) climate model and comparison with the Goddard
24 Institute for Space Studies ModelE. *Atmospheric Chemistry and Physics*, **10**, 5999-6006.
- 25 Jones, R. H., S. Westra, and A. Sharma, 2010b: Observed relationships between extreme sub-daily
26 precipitation, surface temperature, and relative humidity. *Geophysical Research Letters*, **37**.
- 27 Jury, M. R., and M. J. Santiago, 2010: Composite analysis of dust impacts on African easterly waves in the
28 Moderate Resolution Imaging Spectrometer era. *Journal of Geophysical Research-Atmospheres*, **115**,
29 11.
- 30 Kahn, R. A., B. J. Gaitley, J. V. Martonchik, D. J. Diner, K. A. Crean, and B. Holben, 2005: Multiangle
31 Imaging Spectroradiometer (MISR) global aerosol optical depth validation based on 2 years of
32 coincident Aerosol Robotic Network (AERONET) observations. *Journal of Geophysical Research*,
33 **110**, D10s04.
- 34 Kahn, R. A., and Coauthors, 2007: Satellite-derived aerosol optical depth over dark water from MISR and
35 MODIS: Comparisons with AERONET and implications for climatological studies. *Journal of*
36 *Geophysical Research*, **112**, D18205.
- 37 Kanakidou, M., and Coauthors, 2005: Organic aerosol and global climate modelling: a review. *Atmospheric*
38 *Chemistry and Physics*, **5**, 1053-1123.
- 39 Kanji, Z. A., and J. P. D. Abbatt, 2006: Laboratory studies of ice formation via deposition mode nucleation
40 onto mineral dust and n-hexane soot samples. *Journal of Geophysical Research*, **111**, D16204.
- 41 Kanji, Z. A., O. Florea, and J. P. D. Abbatt, 2008: Ice formation via deposition nucleation on mineral dust
42 and organics: dependence of onset relative humidity on total particulate surface area. *Environmental*
43 *Research Letters*, **3**, 025004.
- 44 Kanji, Z. A., P. J. DeMott, O. Mohler, and J. P. D. Abbatt, 2011: Results from the University of Toronto
45 continuous flow diffusion chamber at ICIS 2007: instrument intercomparison and ice onsets for
46 different aerosol types. *Atmospheric Chemistry and Physics*, **11**, 31-41.
- 47 Kärcher, B., and U. Lohmann, 2003: A parameterization of cirrus cloud formation: Heterogeneous freezing.
48 *Journal of Geophysical Research*, **108**, 4402.
- 49 Kärcher, B., and J. Strom, 2003: The roles of dynamical variability and aerosols in cirrus cloud formation.
50 *Atmospheric Chemistry and Physics*, **3**, 823-838.
- 51 Kärcher, B., U. Burkhardt, M. Ponater, and C. Frömming, 2010: Importance of representing optical depth
52 variability for estimates of global line-shaped contrail radiative forcing *Proc. National Academy of*
53 *Science of the USA*, **107**, 19181-19184.
- 54 Kärcher, B., O. Mohler, P. J. DeMott, S. Pechtl, and F. Yu, 2007: Insights into the role of soot aerosols in
55 cirrus cloud formation. *Atmospheric Chemistry and Physics*, **7**, 4203-4227.

- 1 Karl, M., A. Gross, C. Leck, and L. Pirjola, 2007: Intercomparison of dimethylsulfide oxidation mechanisms
2 for the marine boundary layer: Gaseous and particulate sulfur constituents. *Journal of Geophysical*
3 *Research*, **112**, ARTN D15304.
- 4 Kaufman, Y. J., and M. D. Chou, 1993: Model simulations of the competing climatic effects of SO₂ and
5 CO₂. *Journal of Climate*, **6**, 1241-1252.
- 6 Kay, J. E., and A. Gettelman, 2009: Cloud influence on and response to seasonal Arctic sea ice loss. *Journal*
7 *of Geophysical Research*, **114**, D18204.
- 8 Kay, J. E., K. Raeder, A. Gettelman, and J. Anderson, 2011: The boundary layer response to recent Arctic
9 sea ice loss and implications for high-latitude climate feedbacks. *Journal of Climate*, **24**, 428-447.
- 10 Kazil, J., R. G. Harrison, and E. R. Lovejoy, 2008: Tropospheric new particle formation and the role of ions.
11 *Space Science Reviews*, **137**, 241-255.
- 12 Kazil, J., E. Lovejoy, M. Barth, and K. O'Brien, 2006: Aerosol nucleation over oceans and the role of
13 galactic cosmic rays. *Atmospheric Chemistry and Physics*, **6**, 4905-4924.
- 14 Kazil, J., H. Wang, G. Feingold, A. D. Clarke, J. R. Snider, and A. R. Bandy, 2011: Modeling chemical and
15 aerosol processes in the transition from closed to open cells during VOCALS-REx. *Atmospheric*
16 *Chemistry and Physics*, **11**, 7491-7514.
- 17 Kazil, J., and Coauthors, 2010: Aerosol nucleation and its role for clouds and Earth's radiative forcing in the
18 aerosol-climate model ECHAM5-HAM. *Atmospheric Chemistry and Physics*, **10**, 10733-10752.
- 19 Keene, W. C., and Coauthors, 2007: Chemical and physical characteristics of nascent aerosols produced by
20 bursting bubbles at a model air-sea interface. *Journal of Geophysical Research*, **112**, ARTN D21202.
- 21 Keith, D. W., 2000: Geoengineering the climate: History and prospect. *Annual Review of Energy*
22 *Environment*, **25**, 245-284.
- 23 ———, 2010: Photophoretic levitation of engineered aerosols for geoengineering. *Proceedings of the National*
24 *Academy of Sciences of the United States of America*, **107**, 16428-16431.
- 25 Kerkweg, A., J. Buchholz, L. Ganzeveld, A. Pozzer, H. Tost, and P. Jöckel, 2006: Technical Note: An
26 implementation of the dry removal processes DRY DEPosition and SEDimentation in the Modular
27 Earth Submodel System (MESSy). *Atmospheric Chemistry and Physics*, **6**, 4617-4632.
- 28 Kerminen, V. M., and Coauthors, 2010: Atmospheric nucleation: highlights of the EUCAARI project and
29 future directions. *Atmospheric Chemistry and Physics*, **10**, 10829-10848.
- 30 Kernthaler, S. C., R. Toumi, and J. D. Haigh, 1999: Some doubts concerning a link between cosmic ray
31 fluxes and global cloudiness. *Geophysical Research Letters*, **26**, 863-865.
- 32 Khain, A., D. Rosenfeld, and A. Pokrovsky, 2005: Aerosol impact on the dynamics and microphysics of
33 deep convective clouds. *Quarterly Journal of the Royal Meteorological Society*, **131**, 2639-2663.
- 34 Khain, A. P., 2009: Notes on state-of-the-art investigations of aerosol effects on precipitation: a critical
35 review. *Environmental Research Letters*, **4**, ARTN 015004.
- 36 Khairoutdinov, M., D. Randall, and C. DeMott, 2005: Simulations of the atmospheric general circulation
37 using a cloud-resolving model as a superparameterization of physical processes. *Journal of the*
38 *Atmospheric Sciences*, **62**, 2136-2154.
- 39 Khairoutdinov, M. F., and D. A. Randall, 2001: A cloud resolving model as a cloud parameterization in the
40 NCAR Community Climate System Model: Preliminary results. *Geophysical Research Letters*, **28**,
41 3617-3620.
- 42 Khairoutdinov, M. F., S. K. Krueger, C.-H. Moeng, P. A. Bogenschutz, and D. A. Randall, 2009: Large-eddy
43 simulation of maritime deep tropical convection. *Journal of Advances in Modeling Earth Systems*, **1**,
44 15.
- 45 Khan, M. F., Y. Shirasuna, K. Hirano, and S. Masunaga, 2010: Characterization of PM_{2.5}, PM_{2.5-10} and
46 PM₁₀ in ambient air, Yokohama, Japan. *Atmospheric Research*, **96**, 159-172.
- 47 Khare, P., and B. P. Baruah, 2010: Elemental characterization and source identification of PM_{2.5} using
48 multivariate analysis at the suburban site of North-East India. *Atmospheric Research*, **98**, 148-162.
- 49 Khvorostyanov, V., and K. Sassen, 1998: Toward the theory of homogeneous nucleation and its
50 parameterization for cloud models. *Geophysical Research Letters*, **25**, 3155-3158.
- 51 Khvorostyanov, V.I., and J.A. Curry, 2009: Critical humidities of homogeneous and heterogeneous ice
52 nucleation: Inferences from extended classical nucleation theory. *Journal of the Geophysical*
53 *Research*, **114**, Do4207, doi:10.1029/2008JD011197.
- 54 Khvorostyanov, V. I., and J. A. Curry, 2005: Fall velocities of hydrometeors in the atmosphere: Refinements
55 to a continuous analytical power law. *Journal of the Atmospheric Sciences*, **62**, 4343-4357.
- 56 Kiehl, J. T., 1994: On the observed near cancellation between longwave and shortwave cloud forcing in
57 tropical regions. *Journal of Climate*, **7**, 559-565.

- 1 Kiehl, J. T., 2007: Twentieth century climate model response and climate sensitivity. *Geophysical Research*
2 *Letters*, **34**, L22710.
- 3 Kiehl, J. T., T. L. Schneider, P. J. Rasch, M. C. Barth, and J. Wong, 2000: Radiative forcing due to sulfate
4 aerosols from simulations with the National Center for Atmospheric Research Community Climate
5 Model, Version 3. *Journal of Geophysical Research*, **105**, 1441-1457.
- 6 Kim, B. M., S. Teffera, and M. D. Zeldin, 2000: Characterization of PM_{2.5} and PM₁₀ in the South Coast air
7 basin of Southern California: Part 1- Spatial variations. *Journal of the Air & Waste Management*
8 *Association*, **50**, 2034-2044.
- 9 Kim, H.-S., J.-B. Huh, P. K. Hopke, T. M. Holsen, and S.-M. Yi, 2007: Characteristics of the major chemical
10 constituents of PM_{2.5} and smog events in Seoul, Korea in 2003 and 2004. *Atmospheric Environment*,
11 **41**, 6762-6770.
- 12 Kim, J. M., and Coauthors, 2010: Enhanced production of oceanic dimethylsulfide resulting from CO₂-
13 induced grazing activity in a high CO₂ world. *Environmental Science & Technology*, **44**, 8140-8143.
- 14 King, S. M., and Coauthors, 2010: Cloud droplet activation of mixed organic-sulfate particles produced by
15 the photooxidation of isoprene. *Atmospheric Chemistry and Physics*, **10**, 3953-3964.
- 16 Kinne, S., and Coauthors, 2006: An AeroCom initial assessment - optical properties in aerosol component
17 modules of global models. *Atmospheric Chemistry and Physics*, **6**, 1815-1834.
- 18 Kirchstetter, T. W., T. Novakov, and P. V. Hobbs, 2004: Evidence that the spectral dependence of light
19 absorption by aerosols is affected by organic carbon. *Journal of Geophysical Research*, **109**, D21208.
- 20 Kirkby, J., 2007: Cosmic rays and climate. *Surveys in Geophysics*, **28**, 333-375.
- 21 Kirkby, J., and Coauthors, 2011: Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric
22 aerosol nucleation. *Nature*, **476**, 429-433.
- 23 Kirkevåg, A., T. Iversen, O. Seland, J. B. Debernard, T. Storelvmo, and J. E. Kristjansson, 2008: Aerosol-
24 cloud-climate interactions in the climate model CAM-Oslo. *Tellus Series a-Dynamic Meteorology and*
25 *Oceanography*, **60**, 492-512.
- 26 Kleeman, M. J., 2008: A preliminary assessment of the sensitivity of air quality in California to global
27 change. *Climatic Change*, **87**, S273-S292.
- 28 Kleidman, R. G., A. Smirnov, R. C. Levy, S. Mattoo, and D. Tanré, 2011: Evaluation and wind speed
29 dependence of MODIS aerosol retrievals over open ocean. *IEEE Transactions on Geoscience and*
30 *Remote Sensing*, **PP**, 1-7.
- 31 Klein, R. J. T., S. Huq, F. Denton, T. E. Downing, R. G. Richels, J. B. Robinson, and F. L. Toth, 2007: Inter-
32 relationships between adaptation and mitigation. Climate Change 2007: Impacts, Adaptation and
33 Vulnerability. Contribution of Working Group II to the Fourth Assessment Report of the
34 Intergovernmental Panel on Climate Change., 745-777 pp.
- 35 Klein, S. A., and D. L. Hartmann, 1993: The seasonal cycle of low stratiform clouds. *Journal of Climate*, **6**,
36 1587-1606.
- 37 Klein, S. A., and Coauthors, 2009: Intercomparison of model simulations of mixed-phase clouds observed
38 during the ARM Mixed-Phase Arctic Cloud Experiment. I: Single-layer cloud. *Quarterly Journal of*
39 *the Royal Meteorological Society*, **135**, 979-1002.
- 40 Kloster, S., and Coauthors, 2010: Fire dynamics during the 20th century simulated by the Community Land
41 Model. *Biogeosciences*, **7**, 1877-1902.
- 42 Knopf, D. A., and T. Koop, 2006: Heterogeneous nucleation of ice on surrogates of mineral dust. *Journal of*
43 *Geophysical Research*, **111**, D12201.
- 44 Knopf, D. A., P. A. Alpert, B. Wang, and J. Y. Aller, 2011: Stimulation of ice nucleation by marine diatoms.
45 *Nature Geoscience*, **4**, 88-90.
- 46 Knutti, R., T. F. Stocker, F. Joos, and G. K. Plattner, 2002: Constraints on radiative forcing and future
47 climate change from observations and climate model ensembles. *Nature*, **416**, 719-723.
- 48 Kocak, M., N. Mihalopoulos, and N. Kubilay, 2007: Chemical composition of the fine and coarse fraction of
49 aerosols in the northeastern Mediterranean. *Atmospheric Environment*, **41**, 7351-7368.
- 50 Koch, D., and A. D. Del Genio, 2010: Black carbon semi-direct effects on cloud cover: review and synthesis.
51 *Atmospheric Chemistry and Physics*, **10**, 7685-7696.
- 52 Koch, D., S. Menon, A. D. Genio, R. Ruedy, I. Alienov, and G. A. Schmidt, 2009a: Distinguishing aerosol
53 impacts on climate over the past century. *Journal of Climate*, **22**, 2659-2677.
- 54 Koch, D., and Coauthors, 2009b: Evaluation of black carbon estimations in global aerosol models.
55 *Atmospheric Chemistry and Physics*, **9**, 9001-9026.

- 1 Koehler, K. A., S. M. Kreidenweis, P. J. DeMott, M. D. Petters, A. J. Prenni, and O. Mohler, 2010:
2 Laboratory investigations of the impact of mineral dust aerosol on cold cloud formation. *Atmospheric*
3 *Chemistry and Physics*, **10**, 11955-11968.
- 4 Koehler, K. A., and Coauthors, 2009: Cloud condensation nuclei and ice nucleation activity of hydrophobic
5 and hydrophilic soot particles. *Physical Chemistry Chemical Physics*, **11**, 7906-7920.
- 6 Koffi, B., and Coauthors, 2011: Application of the CALIOP Layer Product to evaluate the vertical
7 distribution of aerosols estimated by global models: Part 1. AeroCom phase I results, submitted.
- 8 Köhler, M., M. Ahlgrimm, and A. Beljaars, 2011: Unified treatment of dry convective and stratocumulus-
9 topped boundary layers in the ECMWF model. *Quarterly Journal of the Royal Meteorological*
10 *Society*, **137**, 43-57.
- 11 Kok, J. F., 2011: A scaling theory for the size distribution of emitted dust aerosols suggests climate models
12 underestimate the size of the global dust cycle. *Proceedings of the National Academy of Sciences of*
13 *the United States of America*, **108**, 1016-1021.
- 14 Kokhanovsky, A. A., and Coauthors, 2010: The inter-comparison of major satellite aerosol retrieval
15 algorithms using simulated intensity and polarization characteristics of reflected light. *Atmospheric*
16 *Measurement Techniques*, **3**, 909-932.
- 17 Kondo, Y., and Coauthors, 2011: Emissions of black carbon, organic, and inorganic aerosols from biomass
18 burning in North America and Asia in 2008. *Journal of Geophysical Research*, **116**, D08204.
- 19 Koop, T., B. P. Luo, A. Tsias, and T. Peter, 2000: Water activity as the determinant for homogeneous ice
20 nucleation in aqueous solutions. *Nature*, **406**, 611-614.
- 21 Koren, I., G. Feingold, and L. A. Remer, 2010: The invigoration of deep convective clouds over the Atlantic:
22 aerosol effect, meteorology or retrieval artifact? *Atmospheric Chemistry and Physics*, **10**, 8855-8872.
- 23 Koren, I., Y. Kaufman, L. Remer, and J. Martins, 2004: Measurement of the effect of Amazon smoke on
24 inhibition of cloud formation. *Science*, **303**, 1342-1345.
- 25 Koren, I., J. V. Martins, L. A. Remer, and H. Afargan, 2008: Smoke invigoration versus inhibition of clouds
26 over the Amazon. *Science*, **321**, 946-949.
- 27 Koren, I., Y. J. Kaufman, D. Rosenfeld, L. A. Remer, and Y. Rudich, 2005: Aerosol invigoration and
28 restructuring of Atlantic convective clouds. *Geophysical Research Letters*, **32**, L14828.
- 29 Koren, I., L. A. Remer, Y. J. Kaufman, Y. Rudich, and J. V. Martins, 2007: On the twilight zone between
30 clouds and aerosols. *Geophysical Research Letters*, **34**, L08805.
- 31 Korhonen, H., K. S. Carslaw, and S. Romakkaniemi, 2010a: Enhancement of marine cloud albedo via
32 controlled sea spray injections: a global model study of the influence of emission rates, microphysics
33 and transport. *Atmospheric Chemistry and Physics*, 4133-4143.
- 34 Korhonen, H., K. S. Carslaw, D. V. Spracklen, G. W. Mann, and M. T. Woodhouse, 2008: Influence of
35 oceanic dimethyl sulfide emissions on cloud condensation nuclei concentrations and seasonality over
36 the remote Southern Hemisphere oceans: A global model study. *Journal of Geophysical Research*,
37 **113**, ARTN D15204.
- 38 Korhonen, H., K. S. Carslaw, P. M. Forster, S. Mikkonen, N. D. Gordon, and H. Kokkola, 2010b: Aerosol
39 climate feedback due to decadal increases in Southern Hemisphere wind speeds. *Geophysical*
40 *Research Letters*, ARTN L02805.
- 41 Korolev, A., 2007: Limitations of the Wegener-Bergeron-Findeisen mechanism in the evolution of mixed-
42 phase clouds. *Journal of the Atmospheric Sciences*, **64**, 3372-3375.
- 43 Korolev, A., and P. R. Field, 2008: The effect of dynamics on mixed-phase clouds: Theoretical
44 considerations. *Journal of the Atmospheric Sciences*, **65**, 66-86.
- 45 Kostinski, A. B., 2008: Drizzle rates versus cloud depths for marine stratocumuli. *Environmental Research*
46 *Letters*, **3**, 045019.
- 47 Kravitz, B., A. Robock, O. Boucher, H. Schmidt, K. Talyor, G. Stinchikov, and M. Schulz, 2011: The
48 Geoengineering Model Intercomparison Project (GeoMIP). *Atmospheric Science Letters*, **12**, 162-167.
- 49 Kristjánsson, J. E., 2002: Studies of the aerosol indirect effect from sulfate and black carbon aerosols.
50 *Journal of Geophysical Research*, **107**, 4246.
- 51 Kristjánsson, J. E., T. Iversen, A. Kirkevåg, O. Seland, and J. Debernard, 2005: Response of the climate
52 system to aerosol direct and indirect forcing: Role of cloud feedbacks. *Journal of Geophysical*
53 *Research*, **110**, D24206.
- 54 Kristjánsson, J. E., C. W. Stjern, F. Stordal, A. M. Fjæraa, G. Myhre, and K. Jónasson, 2008: Cosmic rays,
55 cloud condensation nuclei and clouds – a reassessment using MODIS data. *Atmospheric Chemistry*
56 *and Physics*, **8**, 7373-7387.

- 1 Kroll, J. H., and J. H. Seinfeld, 2008: Chemistry of secondary organic aerosol: Formation and evolution of
2 low-volatility organics in the atmosphere. *Atmospheric Environment*, **42**, 3593-3624.
- 3 Kuang, C., P. H. McMurry, A. V. McCormick, and F. L. Eisele, 2008: Dependence of nucleation rates on
4 sulfuric acid vapor concentration in diverse atmospheric locations. *Journal of Geophysical Research*,
5 **113**, ARTN D10209.
- 6 Kuang, Z., and D. Hartmann, 2007: Testing the fixed anvil temperature hypothesis in a cloud-resolving
7 model. *Journal of Climate*, **20**, 2051-2057.
- 8 Kubar, T. L., D. L. Hartmann, and R. Wood, 2007: Radiative and convective driving of tropical high clouds.
9 *Journal of Climate*, **20**, 5510-5526.
- 10 Kueppers, L. M., M. A. Snyder, and L. C. Sloan, 2007: Irrigation cooling effect: Regional climate forcing by
11 land-use change. *Geophys. Res. Lett.*, **34**, L03703.
- 12 Kulkarni, G., and S. Dobbie, 2010: Ice nucleation properties of mineral dust particles: determination of onset
13 RH(i), IN active fraction, nucleation time-lag, and the effect of active sites on contact angles.
14 *Atmospheric Chemistry and Physics*, **10**, 95-105.
- 15 Kulmala, M., and V. M. Kerminen, 2008: On the formation and growth of atmospheric nanoparticles.
16 *Atmospheric Research*, **90**, 132-150.
- 17 Kulmala, M., and Coauthors, 2010: Atmospheric data over a solar cycle: no connection between galactic
18 cosmic rays and new particle formation. *Atmospheric Chemistry and Physics*, **10**, 1885-1898.
- 19 Kumar, R., S. S. Srivastava, and K. M. Kumari, 2007: Characteristics of aerosols over suburban and urban
20 site of semiarid region in India: Seasonal and spatial variations. *Aerosol and Air Quality Research*, **7**,
21 531-549.
- 22 Kurten, T., V. Loukonen, H. Vehkamäki, and M. Kulmala, 2008: Amines are likely to enhance neutral and
23 ion-induced sulfuric acid-water nucleation in the atmosphere more effectively than ammonia.
24 *Atmospheric Chemistry and Physics*, **8**, 4095-4103.
- 25 Kvalevag, M. M., and G. Myhre, 2007: Human impact on direct and diffuse solar radiation during the
26 industrial era. *Journal of Climate*, **20**, 4874-4883.
- 27 Laken, B., A. Wolfendale, and D. Kniveton, 2009: Cosmic ray decreases and changes in the liquid water
28 cloud fraction over the oceans. *Geophysical Research Letters*, **36**, L23803.
- 29 Laken, B. A., D. R. Kniveton, and M. R. Frogley, 2010: Cosmic rays linked to rapid mid-latitude cloud
30 changes. *Atmospheric Chemistry and Physics*, **10**, 10941-10948.
- 31 Lamarque, J.-F., and Coauthors, 2010: Historical (1850-2000) gridded anthropogenic and biomass burning
32 emissions of reactive gases and aerosols: methodology and application. *Atmospheric Chemistry and
33 Physics*, **10**, 7017-7039.
- 34 Lambert, F. H., and M. R. Allen, 2009: Are changes in global precipitation constrained by the tropospheric
35 energy budget? *Journal of Climate*, **22**, 499-517.
- 36 Lambert, F. H., M. J. Webb, and M. M. Joshi, 2011: The relationship between land-ocean surface
37 temperature contrast and radiative forcing. *Journal of Climate*, **24**, 3239-3256.
- 38 Lana, A., and Coauthors, 2011: An updated climatology of surface dimethylsulfide concentrations and
39 emission fluxes in the global ocean. *Global Biogeochemical Cycles*, **25**, ARTN GB1004.
- 40 Lance, S., and Coauthors, 2011: Cloud condensation nuclei as a modulator of ice processes in Arctic mixed-
41 phase clouds. *Atmospheric Chemistry and Physics*, **11**, 8003-8015.
- 42 Lanz, V. A., M. R. Alfarra, U. Baltensperger, B. Buchmann, C. Hueglin, and A. S. H. Prévôt, 2007: Source
43 apportionment of submicron organic aerosols at an urban site by factor analytical modelling of aerosol
44 mass spectra. *Atmospheric Chemistry and Physics*, **7**, 1503-1522.
- 45 Latham, J., 1990: Control of global warming? *Nature*, **347**, 339-340.
- 46 Latham, J., and Coauthors, 2008: Global temperature stabilization via controlled albedo enhancement of
47 low-level maritime clouds. *Philosophical Transactions Royal Society London*, **366**, 3969-3987.
- 48 Lathièrè, J., C. N. Hewitt, and D. J. Beerling, 2010: Sensitivity of isoprene emissions from the terrestrial
49 biosphere to 20th century changes in atmospheric CO₂ concentration, climate, and land use. *Global
50 Biogeochemical Cycles*, **24**, GB1004.
- 51 Lau, K. M., and K. M. Kim, 2007: Cooling of the Atlantic by Saharan dust. *Geophys. Res. Lett.*, **34**, L23811.
- 52 Lauer, A., K. Hamilton, Y. Q. Wang, V. T. J. Phillips, and R. Bennartz, 2010: The impact of global warming
53 on marine boundary layer clouds over the Eastern Pacific - A regional model study. *Journal of
54 Climate*, **23**, 5844-5863.
- 55 Le Clainche, Y., and Coauthors, 2010: A first appraisal of prognostic ocean DMS models and prospects for
56 their use in climate models. *Global Biogeochemical Cycles*, **24**, GB3021.

- 1 Lebsock, M. D., G. L. Stephens, and C. Kummerow, 2008: Multisensor satellite observations of aerosol
2 effects on warm clouds. *Journal of Geophysical Research*, **113**, D15205.
- 3 Lee, D., and Coauthors, 2009: Aviation and global climate change in the 21st century. *Atmospheric*
4 *Environment*, 3520-3537.
- 5 Lee, H. S., and B. W. Kang, 2001: Chemical characteristics of principal PM_{2.5} species in Chongju, South
6 Korea. *Atmospheric Environment*, **35**, 739-749.
- 7 Lee, L. A., K. S. Carslaw, K. Pringle, G. W. Mann, and D. V. Spracklen, 2011: Emulation of a complex
8 global aerosol model to quantify sensitivity to uncertain parameters. *Atmospheric Chemistry and*
9 *Physics Discussions*, **11**, 20433-20485.
- 10 Lee, S. S., L. J. Donner, and J. E. Penner, 2010: Thunderstorm and stratocumulus: how does their contrasting
11 morphology affect their interactions with aerosols? *Atmospheric Chemistry and Physics*, **10**, 6819-
12 6837.
- 13 Lenderink, G., and E. Van Meijgaard, 2008: Increase in hourly precipitation extremes beyond expectations
14 from temperature changes. *Nature Geoscience*, **1**, 511-514.
- 15 ———, 2010: Linking increases in hourly precipitation extremes to atmospheric temperature and moisture
16 changes. *Environmental Research Letters*, **5**.
- 17 Lenschow, P., H. J. Abraham, K. Kutzner, M. Lutz, J. D. Preu, and W. Reichenbacher, 2001: Some ideas
18 about the sources of PM₁₀. *Atmospheric Environment*, **35**, 23-33.
- 19 Lenton, T. M., and N. E. Vaughan, 2009: The radiative forcing potential of different climate geoengineering
20 options. *Atmospheric Chemistry and Physics*, **9**, 5539-5561.
- 21 Levin, Z., and W. R. Cotton, 2009: *Aerosol Pollution Impact on Precipitation: A Scientific Review*. Springer
22 Verlag, 386 pp. pp.
- 23 Levy, R. C., L. A. Remer, S. Mattoo, E. F. Vermote, and Y. J. Kaufman, 2007: Second-generation
24 operational algorithm: Retrieval of aerosol properties over land from inversion of Moderate Resolution
25 Imaging Spectroradiometer spectral reflectance. *Journal of Geophysical Research*, **112**, D13211.
- 26 Levy, R. C., L. A. Remer, R. G. Kleidman, S. Mattoo, C. Ichoku, R. Kahn, and T. F. Eck, 2010: Global
27 evaluation of the Collection 5 MODIS dark-target aerosol products over land. *Atmospheric Chemistry*
28 *and Physics*, **10**, 10399-10420.
- 29 Li, J., M. Pósfai, P. V. Hobbs, and P. R. Buseck, 2003: Individual aerosol particles from biomass burning in
30 southern Africa: 2, Compositions and aging of inorganic particles. *Journal of Geophysical Research*,
31 **108**, 8484.
- 32 Li, Z., K.-H. Lee, Y. Wang, J. Xin, and W.-M. Hao, 2010: First observation-based estimates of cloud-free
33 aerosol radiative forcing across China. *Journal of Geophysical Research-Atmospheres*, **115**, D00K18.
- 34 Liao, H., W. T. Chen, and J. H. Seinfeld, 2006: Role of climate change in global predictions of future
35 tropospheric ozone and aerosols. *Journal of Geophysical Research*, **111**, D12304.
- 36 Liao, H., Y. Zhang, W. T. Chen, F. Raes, and J. H. Seinfeld, 2009: Effect of chemistry-aerosol-climate
37 coupling on predictions of future climate and future levels of tropospheric ozone and aerosols. *Journal*
38 *of Geophysical Research*, **114**, D10306.
- 39 Libardoni, A. G., and C. E. Forest, 2011: Sensitivity of distributions of climate system properties to the
40 surface temperature dataset. *Geophysical Research Letters*, online.
- 41 Liepert, B. G., and M. Previdi, 2009: Do models and observations disagree on the rainfall response to global
42 warming? *Journal of Climate*, **22**, 3156-3166.
- 43 Lin, J. C., T. Matsui, R. A. S. Pielke, and C. Kummerow, 2006: Effects of biomass burning-derived aerosols
44 on precipitation and clouds in the Amazon Basin: A satellite-based empirical study. *J. Geophys. Res.*,
45 **111**, D19204.
- 46 Lindsey, D. T., and M. Fromm, 2008: Evidence of the cloud lifetime effect from wildfire-induced
47 thunderstorms. *Geophysical Research Letters*, **35**, L22809.
- 48 Lindzen, R. S., and Y.-S. Choi, 2011: On the observational determination of climate sensitivity and its
49 implications. *Asia-Pacific Journal of Atmospheric Sciences*, **47**, 377-390.
- 50 Lintner, B. R., C. E. Holloway, and J. D. Neelin, 2011: Column Water Vapor Statistics and Their
51 Relationship to Deep Convection, Vertical and Horizontal Circulation, and Moisture Structure at
52 Nauru. *Journal of Climate*, **24**, 5454-5466.
- 53 Liou, K. N., and S. C. Ou, 1989: The role of cloud microphysical processes in climate - An assessment from
54 a one-dimensional perspective. *Journal of Geophysical Research*, **94**, 8599-8607.
- 55 Liu, W., Y. Wang, A. Russell, and E. S. Edgerton, 2005a: Atmospheric aerosol over two urban-rural pairs in
56 the southeastern United States: Chemical composition and possible sources. *Atmospheric*
57 *Environment*, **39**, 4453-4470.

- 1 Liu, X., J. Penner, S. Ghan, and M. Wang, 2007: Inclusion of ice microphysics in the NCAR community
2 atmospheric model version 3 (CAM3). *Journal of Climate*, 4526-4547.
- 3 Liu, X. H., J. E. Penner, and M. Herzog, 2005b: Global modeling of aerosol dynamics: Model description,
4 evaluation, and interactions between sulfate and nonsulfate aerosols. *Journal of Geophysical*
5 *Research*, D18206.
- 6 Liu, X. H., J. E. Penner, and M. H. Wang, 2009: Influence of anthropogenic sulfate and black carbon on
7 upper tropospheric clouds in the NCAR CAM3 model coupled to the IMPACT global aerosol model.
8 *Journal of Geophysical Research*, **114**, D03204.
- 9 Liu, Y., and P. H. Daum, 2002: Anthropogenic aerosols: Indirect warming effect from dispersion forcing.
10 *Nature*, **419**, 580-581.
- 11 Liu, Y., J. R. Key, and X. Wang, 2008: The influence of changes in cloud cover on recent surface
12 temperature trends in the Arctic. *Journal of Climate*, **21**, 705-715.
- 13 Lobell, D., G. Bala, A. Mirin, T. Phillips, R. Maxwell, and D. Rotman, 2009: Regional differences in the
14 influence of irrigation on climate. *J. Climate*, **22**, 2248–2255.
- 15 Lodhi, A., B. Ghauri, M. R. Khan, S. Rahmana, and S. Shafiquea, 2009: Particulate matter (PM_{2.5})
16 concentration and source apportionment in Lahore. *Journal of the Brazilian Chemical Society*, **20**,
17 1811-1820.
- 18 Loeb, N. G., and N. Manalo-Smith, 2005: Top-of-atmosphere direct radiative effect of aerosols over global
19 oceans from merged CERES and MODIS observations. *Journal of Climate*, **18**, 3506-3526.
- 20 Loeb, N. G., and W. Y. Su, 2010: Direct aerosol radiative forcing uncertainty based on a radiative
21 perturbation analysis. *Journal of Climate*, **23**, 5288-5293.
- 22 Loeb, N. G., and Coauthors, 2009: Toward optimal closure of the Earth's top-of-atmosphere radiation
23 budget. *Journal of Climate*, **22**, 748-766.
- 24 Lohmann, U., 2002a: A glaciation indirect aerosol effect caused by soot aerosols. *Geophysical Research*
25 *Letters*, **29**, 1052.
- 26 ———, 2002b: Possible aerosol effects on ice clouds via contact nucleation. *Journal of the Atmospheric*
27 *Sciences*, **59**, 647-656.
- 28 ———, 2004: Can anthropogenic aerosols decrease the snowfall rate? *Journal of the Atmospheric Sciences*,
29 **61**, 2457-2468.
- 30 ———, 2008: Global anthropogenic aerosol effects on convective clouds in ECHAM5-HAM. *Atmospheric*
31 *Chemistry and Physics*, **8**, 2115-2131.
- 32 Lohmann, U., and J. Feichter, 1997: Impact of sulfate aerosols on albedo and lifetime of clouds: A sensitivity
33 study with the ECHAM4 GCM. *Journal of Geophysical Research*, **102**, 13685-13700.
- 34 ———, 2001: Can the direct and semi-direct aerosol effect compete with the indirect effect on a global scale?
35 *Geophysical Research Letters*, **28**, 159-161.
- 36 Lohmann, U., and G. Lesins, 2002: Stronger constraints on the anthropogenic indirect aerosol effect.
37 *Science*, **298**, 1012-1015.
- 38 Lohmann, U., and B. Kärcher, 2002: First interactive simulations of cirrus clouds formed by homogeneous
39 freezing in the ECHAM general circulation model. *Journal of Geophysical Research*, **107**, 4105.
- 40 Lohmann, U., and J. Feichter, 2005: Global indirect aerosol effects: a review. *Atmospheric Chemistry and*
41 *Physics*, **5**, 715-737.
- 42 Lohmann, U., and K. Diehl, 2006: Sensitivity studies of the importance of dust ice nuclei for the indirect
43 aerosol effect on stratiform mixed-phase clouds. *Journal of the Atmospheric Sciences*, **63**, 968-982.
- 44 Lohmann, U., and C. Hoose, 2009: Sensitivity studies of different aerosol indirect effects in mixed-phase
45 clouds. *Atmospheric Chemistry and Physics*, **9**, 8917-8934.
- 46 Lohmann, U., and S. Ferrachat, 2010: Impact of parametric uncertainties on the present-day climate and on
47 the anthropogenic aerosol effect. *Atmospheric Chemistry and Physics*, **10**, 11373-11383.
- 48 Lohmann, U., J. Feichter, J. Penner, and R. Leaitch, 2000: Indirect effect of sulfate and carbonaceous
49 aerosols: A mechanistic treatment. *Journal of Geophysical Research*, **105**, 12193-12206.
- 50 Lohmann, U., P. Stier, C. Hoose, S. Ferrachat, S. Kloster, E. Roeckner, and J. Zhang, 2007: Cloud
51 microphysics and aerosol indirect effects in the global climate model ECHAM5-HAM. *Atmospheric*
52 *Chemistry and Physics*, **7**, 3425-3446.
- 53 Lohmann, U., and Coauthors, 2010: Total aerosol effect: radiative forcing or radiative flux perturbation?
54 *Atmospheric Chemistry and Physics*, **10**, 3235-3246.
- 55 Lonati, G., M. Giugliano, P. Butelli, L. Romele, and R. Tardivo, 2005: Major chemical components of PM_{2.5}
56 in Milan (Italy). *Atmospheric Environment*, **39**, 1925-1934.

- 1 Lu, M.-L., W. C. Conant, H. H. Jonsson, V. Varutbangkul, R. C. Flagan, and J. H. Seinfeld, 2007: The
2 marine stratus/stratocumulus experiment (MASE): aerosol-cloud relationships in marine
3 stratocumulus. *Journal of Geophysical Research*, **112**, D10209.
- 4 Lu, M.-L., G. Feingold, H. H. Jonsson, P. Y. Chuang, H. Gates, R. C. Flagan, and J. H. Seinfeld, 2008:
5 Aerosol-cloud relationships in continental shallow cumulus. *Journal of Geophysical Research*, **113**,
6 D15201.
- 7 Lüönd, F., O. Stetzer, A. Welti, and U. Lohmann, 2010: Experimental study on the ice nucleation ability of
8 size-selected kaolinite particles in the immersion mode. *Journal of Geophysical Research*, **115**,
9 D14201.
- 10 Lynn, B., A. Khain, D. Rosenfeld, and W. L. Woodley, 2007: Effects of aerosols on precipitation from
11 orographic clouds. *Journal of Geophysical Research-Atmospheres*, **112**.
- 12 Lynn, B. H., A. P. Khain, J. Dudhia, D. Rosenfeld, A. Pokrovsky, and A. Seifert, 2005: Spectral (Bin)
13 microphysics coupled with a mesoscale model (MM5). Part II: Simulation of a CaPE rain event with a
14 squall line. *Monthly Weather Review*, **133**, 59-71.
- 15 Maenhaut, W., I. Salma, and J. Cafreyer, 1996: Regional atmospheric aerosol composition and sources in
16 the eastern Transvaal, South Africa, and impact of biomass burning. *Journal of Geophysical Research*,
17 **101**, 23613-23650.
- 18 Maenhaut, W., M.-T. Fernandez-Jimenez, J. L. Vanderzalm, B. Hooper, M. A. Hooper, and N. J. Tapper,
19 2000: Aerosol composition at Jabiru, Australia, and impact of biomass burning. *Journal of Aerosol*
20 *Science*, **31**, 745-746.
- 21 Magee, N., A. M. Moyle, and D. Lamb, 2006: Experimental determination of the deposition coefficient of
22 small cirrus-like ice crystals near -50°C. *Geophysical Research Letters*, **33**, L17813.
- 23 Mahowald, N. M., 2007: Anthropocene changes in desert area: Sensitivity to climate model predictions.
24 *Geophysical Research Letters*, **34**, L18817.
- 25 Mahowald, N. M., and C. Luo, 2003: A less dusty future? *Geophysical Research Letters*, **30**, 1903.
- 26 Mahowald, N. M., J. F. Lamarque, X. X. Tie, and E. Wolff, 2006a: Sea-salt aerosol response to climate
27 change: Last Glacial Maximum, preindustrial, and doubled carbon dioxide climates. *Journal of*
28 *Geophysical Research*, **111**, D05303.
- 29 Mahowald, N. M., D. R. Muhs, S. Levis, P. J. Rasch, M. Yoshioka, C. S. Zender, and C. Luo, 2006b:
30 Change in atmospheric mineral aerosols in response to climate: Last glacial period, preindustrial,
31 modern, and doubled carbon dioxide climates. *Journal of Geophysical Research*, **111**, D10202.
- 32 Mahowald, N. M., and Coauthors, 2009: Atmospheric iron deposition: Global distribution, variability, and
33 human perturbations. *Annual Review of Marine Science*, **1**, 245-278.
- 34 Mahowald, N. M., and Coauthors, 2010: Observed 20th century desert dust variability: impact on climate
35 and biogeochemistry. *Atmospheric Chemistry and Physics*, **10**, 10875-10893.
- 36 Makkonen, R., A. Asmi, V.-M. Kerminen, M. Boy, A. Arneth, P. Hari, and M. Kulmala, 2011: Air pollution
37 control and decreasing new particle formation lead to strong climate warming. *Atmospheric Chemistry*
38 *and Physics Discussion*, **11**, 25991-26007.
- 39 Malm, W. C., and B. A. Schichtel, 2004: Spatial and monthly trends in speciated fine particle concentration
40 in the United States. *Journal of Geophysical Research*, **109**, D03306.
- 41 Malm, W. C., J. F. Sisler, D. Huffman, R. A. Eldred, and T. A. Cahill, 1994: Spatial and seasonal trends in
42 particle concentration and optical extinction in the United States. *Journal of Geophysical Research*,
43 **99**, 1347-1370.
- 44 Mangold, A., and Coauthors, 2005: Experimental investigation of ice nucleation by different types of
45 aerosols in the aerosol chamber AIDA: implications to microphysics of cirrus clouds. *Meteorologische*
46 *Zeitschrift*, **14**, 485-497.
- 47 Mann, G. W., and Coauthors, 2010: Description and evaluation of GLOMAP-mode: a modal global aerosol
48 microphysics model for the UKCA composition-climate model. *Geoscientific Model Development*, **3**,
49 519-551.
- 50 Manninen, H. E., and Coauthors, 2010: EUCAARI ion spectrometer measurements at 12 European sites -
51 analysis of new particle formation events. *Atmospheric Chemistry and Physics*, **10**, 7907-7927.
- 52 Mapes, B., J. Bacmeister, M. Khairoutdinov, C. Hannay, and M. Zhao, 2009: Virtual Field Campaigns on
53 Deep Tropical Convection in Climate Models. *Journal of Climate*, **22**, 244-257.
- 54 Marchand, R., T. Ackerman, M. Smyth, and W. B. Rossow, 2010: A review of cloud top height and optical
55 depth histograms from MISR, ISCCP, and MODIS. *Journal of Geophysical Research*, **115**, D16206.

- 1 Mariani, R. L., and W. Z. d. Mello, 2007: PM_{2.5}–10, PM_{2.5} and associated water-soluble inorganic species
2 at a coastal urban site in the metropolitan region of Rio de Janeiro. *Atmospheric Environment*, **41**,
3 2887–2892.
- 4 Markowicz, K. M., and M. L. Witek, 2011: Simulations of contrail optical properties and radiative forcing
5 for various crystal shapes. *J. Appl. Meteor. Climatol.*, **50**, 1740–1755.
- 6 Marlon, J. R., and Coauthors, 2008: Climate and human influences on global biomass burning over the past
7 two millennia. *Nature Geoscience*, **1**, 697–702.
- 8 Marsh, N. D., and H. Svensmark, 2000: Low cloud properties influenced by cosmic rays. *Physical Review*
9 *Letters*, **85**, 5004–5007.
- 10 Martin, S. T., and Coauthors, 2010: Sources and properties of Amazonian aerosol particles. *Review of*
11 *Geophysics*, **48**, RG2002.
- 12 Martins, J. A., M. A. F. S. Dias, and F. L. T. Goncalves, 2009: Impact of biomass burning aerosols on
13 precipitation in the Amazon: A modeling case study. *Journal of Geophysical Research-Atmospheres*,
14 **114**.
- 15 Matheou, G., D. Chung, L. Nuijens, B. Stevens, and J. Teixeira, 2011: On the fidelity of large-eddy
16 simulation of shallow precipitating cumulus convection. *Monthly Weather Review*, **139**, 2918–2939.
- 17 Mauger, G. S., and J. R. Norris, 2010: Assessing the impact of meteorological history on subtropical cloud
18 fraction. *Journal of Climate*, **23**, 2926–2940.
- 19 May, P. T., J. H. Mather, G. Vaughan, C. Jakob, G. M. McFarquhar, K. N. Bower, and G. G. Mace, 2008:
20 The tropical warm pool international cloud experiment. *Bulletin of the American Meteorological*
21 *Society*, **89**, 629–645.
- 22 McComiskey, A., and G. Feingold, 2008: Quantifying error in the radiative forcing of the first aerosol
23 indirect effect. *Geophysical Research Letters*, **35**, L02810.
- 24 McComiskey, A., and G. Feingold, 2011: The scale problem in quantifying aerosol indirect effects. *Atmos.*
25 *Chem. Phys. Discuss.*, **11**, 26741–26789.
- 26 McConnell, J. R., and Coauthors, 2007: 20th-century industrial black carbon emissions altered arctic climate
27 forcing. *Science*, **317**, 1381–1384.
- 28 McCracken, K. G., and J. Beer, 2007: Long-term changes in the cosmic ray intensity at Earth, 1428–2005. *J.*
29 *Geophys. Res.*, **112**, A10101.
- 30 McFarquhar, G. M., and Coauthors, 2011: Indirect and semi-direct aerosol campaign: The impact of Arctic
31 aerosols on clouds. *Bulletin of the American Meteorological Society*, **92**, 183–201.
- 32 McMeeking, G. R., and Coauthors, 2010: Black carbon measurements in the boundary layer over western
33 and northern Europe. *Atmospheric Chemistry and Physics*, **10**, 9393–9414.
- 34 Medeiros, B., B. Stevens, I. M. Held, M. Zhao, D. L. Williamson, J. G. Olson, and C. S. Bretherton, 2008:
35 Aquaplanets, climate sensitivity, and low clouds. *Journal of Climate*, **21**, 4974–4991.
- 36 Meehl, G. A., J. M. Arblaster, K. Matthes, F. Sassi, and H. van Loon, 2009: Amplifying the Pacific climate
37 system response to a small 11-year solar cycle forcing. *Science*, **325**, 1114–1118.
- 38 Meehl, G. A., and Coauthors, 2007: Global Climate Projections. *Climate Change 2007: The Physical*
39 *Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the*
40 *Intergovernmental Panel on Climate Change*, Cambridge University Press.
- 41 Menon, S., and L. Rotstayn, 2006: The radiative influence of aerosol effects on liquid-phase cumulus and
42 stratiform clouds based on sensitivity studies with two climate models. *Climate Dynamics*, **27**, 345–
43 356.
- 44 Menon, S., and A. DelGenio, 2007: Evaluating the impacts of carbonaceous aerosols on clouds and climate.
45 *Human-Induced Climate Change: An Interdisciplinary Assessment*, S. M. E., H. S. Kheshgi, J. Smith,
46 F. C. de la Chesnaye, J. M. Reilly, T. Wilson, and C. Kolstad, Eds., Cambridge University Press.
- 47 Menon, S., A. D. Del Genio, D. Koch, and G. Tselioudis, 2002: GCM Simulations of the aerosol indirect
48 effect: Sensitivity to cloud parameterization and aerosol burden. *Journal of the Atmospheric Sciences*,
49 **59**, 692–713.
- 50 Mercado, L. M., N. Bellouin, S. Sitch, O. Boucher, C. Huntingford, M. Wild, and P. M. Cox, 2009: Impact
51 of changes in diffuse radiation on the global land carbon sink. *Nature*, **458**, 1014–1017.
- 52 Merikanto, J., D. V. Spracklen, G. W. Mann, S. J. Pickering, and K. S. Carslaw, 2009: Impact of nucleation
53 on global CCN. *Atmospheric Chemistry and Physics*, **9**, 8601–8616.
- 54 Metzger, A., and Coauthors, 2010: Evidence for the role of organics in aerosol particle formation under
55 atmospheric conditions. *Proceedings of the National Academy of Sciences of the United States of*
56 *America*, **107**, 6646–6651.

- 1 Meyers, M. P., P. J. Demott, and W. R. Cotton, 1992: New primary ice-nucleation parametrizations in an
2 explicit cloud model. *Journal of Applied Meteorology*, **31**, 708-721.
- 3 Ming, J., D. Zhang, S. Kang, and W. Tian, 2007a: Aerosol and fresh snow chemistry in the East Rongbuk
4 Glacier on the northern slope of Mt. Qomolangma (Everest). *Journal of Geophysical Research*, **112**,
5 D15307.
- 6 Ming, Y., V. Ramaswamy, P. A. Ginoux, L. W. Horowitz, and L. M. Russell, 2005: Geophysical Fluid
7 Dynamics Laboratory general circulation model investigation of the indirect radiative effects of
8 anthropogenic sulfate aerosol. *Journal of Geophysical Research*, **110**, D22206.
- 9 Ming, Y., V. Ramaswamy, L. J. Donner, V. T. J. Phillips, S. A. Klein, P. A. Ginoux, and L. W. Horowitz,
10 2007b: Modeling the interactions between aerosols and liquid water clouds with a self-consistent
11 cloud scheme in a general circulation model. *Journal of the Atmospheric Sciences*, **64**, 1189-1209.
- 12 Mirme, S., A. Mirme, A. Minikin, A. Petzold, U. Horrak, V. M. Kerminen, and M. Kulmala, 2010:
13 Atmospheric sub-3 nm particles at high altitudes. *Atmospheric Chemistry and Physics*, **10**, 437-451.
- 14 Mishchenko, M. I., and Coauthors, 2007: Accurate monitoring of terrestrial aerosols and total solar
15 irradiance - Introducing the glory mission. *Bulletin of the American Meteorological Society*, **88**, 677-
16 691.
- 17 Mitchell, D. L., and W. Finnegan, 2009: Modification of cirrus clouds to reduce global warming.
18 *Environmental Research Letters*, **4**, 045102.
- 19 Mitchell, D. L., R. P. d'Entremont, and R. P. Lawson, 2010: Inferring Cirrus Size Distributions through
20 Satellite Remote Sensing and Microphysical Databases. *Journal of the Atmospheric Sciences*, **67**,
21 1106-1125.
- 22 Miura, H., H. Tomita, T. Nasuno, S. Iga, M. Satoh, and T. Matsuno, 2005: A climate sensitivity test using a
23 global cloud resolving model under an aqua planet condition. *Geophysical Research Letters*, ARTN
24 L19717, DOI 10.1029/2005GL023672.
- 25 Mkoma, S. L., 2008: Physico-chemical characterisation of atmospheric aerosol in Tanzania, with emphasis
26 on the carbonaceous aerosol components and on chemical mass closure, Department of Analytical
27 Chemistry, Ghent University.
- 28 Mkoma, S. L., W. Maenhaut, X. G. Chi, W. Wang, and N. Raes, 2009a: Characterisation of PM₁₀
29 atmospheric aerosols for the wet season 2005 at two sites in East Africa. *Atmospheric Environment*,
30 **43**, 631-639.
- 31 Mkoma, S. L., W. Maenhaut, X. Chi, W. Wang, and N. Raes, 2009b: Chemical composition and mass
32 closure for PM₁₀ aerosols during the 2005 dry season at a rural site in Morogoro, Tanzania. *X-Ray*
33 *Spectrom*, **38**, 293-300.
- 34 Mohler, O., and Coauthors, 2008: Heterogeneous ice nucleation activity of bacteria: new laboratory
35 experiments at simulated cloud conditions. *Biogeosciences*, **5**, 1425-1435.
- 36 Mohler, O., and Coauthors, 2005: Effect of sulfuric acid coating on heterogeneous ice nucleation by soot
37 aerosol particles. *Journal of Geophysical Research*, **110**, D11210.
- 38 Mohler, O., and Coauthors, 2006: Efficiency of the deposition mode ice nucleation on mineral dust particles.
39 *Atmospheric Chemistry and Physics*, **6**, 3007-3021.
- 40 Mohler, O., and Coauthors, 2003: Experimental investigation of homogeneous freezing of sulphuric acid
41 particles in the aerosol chamber AIDA. *Atmospheric Chemistry and Physics*, **3**, 211-223.
- 42 Morales, J. A., D. Pirela, M. G. d. Nava, B. S. d. Borrego, H. Velasquez, and J. Duran, 1998: Inorganic water
43 soluble ions in atmospheric particles over Maracaibo Lake Basin in the western region of Venezuela.
44 *Atmospheric Research*, **46**, 307-320.
- 45 Morrison, H., and A. Gettelman, 2008: A new two-moment bulk stratiform cloud microphysics scheme in
46 the community atmosphere model, version 3 (CAM3). Part I: Description and numerical tests. *Journal*
47 *of Climate*, 3642-3659.
- 48 Morrison, H., and W. W. Grabowski, 2011: Cloud-system resolving model simulations of aerosol indirect
49 effects on tropical deep convection and its thermodynamic environment. *Atmos. Chem. Phys.*, **11**,
50 10503-10523.
- 51 Morrison, H., P. Zuidema, G. M. McFarquhar, A. Bansemer, and A. J. Heymsfield, 2011: Snow
52 microphysical observations in shallow mixed-phase and deep frontal Arctic cloud systems. *Quarterly*
53 *Journal of the Royal Meteorological Society*, **137**, 1589-1601.
- 54 Morrison, H., G. DeBoer, G. Feingold, J. Y. Harrington, M. Shupe, and K. Sulia, 2012: Resilience of
55 persistent Arctic mixed-phase clouds. *Nature Geoscience*, **in press**.
- 56 Mortazavi, R., C. T. Hayes, and P. A. Ariya, 2008: Ice nucleation activity of bacteria isolated from snow
57 compared with organic and inorganic substrates. *Environmental Chemistry*, **5**, 373-381.

- 1 Moteki, N., and Y. Kondo, 2010: Dependence of laser-induced incandescence on physical properties of
2 black carbon aerosols: Measurements and theoretical interpretation. *Aerosol Science and Technology*,
3 **44**, 663-675.
- 4 Moteki, N., Y. Kondo, N. Takegawa, and S. Nakamura, 2009: Directional dependence of thermal emission
5 from nonspherical carbon particles. *Journal of Aerosol Science*, **40**, 790-801.
- 6 Moteki, N., and Coauthors, 2007: Evolution of mixing state of black carbon particles: Aircraft measurements
7 over the western Pacific in March 2004. *Geophysical Research Letters*, L11803.
- 8 Mouillot, F., A. Narasimha, Y. Balkanski, J. F. Lamarque, and C. B. Field, 2006: Global carbon emissions
9 from biomass burning in the 20th century. *Geophysical Research Letters*, **33**, L01801.
- 10 Muhlbauer, A., and U. Lohmann, 2009: Sensitivity studies of aerosol-cloud interactions in mixed-phase
11 orographic precipitation. *Journal of the Atmospheric Sciences*, **66**, 2517-2538.
- 12 Muller, C. J., P. A. O'Gorman, and L. E. Back, 2011: Intensification of precipitation extremes with warming
13 in a cloud-resolving model. *Journal of Climate*, **24**, 2784-2800.
- 14 Muller, G., and A. Chlond, 1996: Three-dimensional numerical study of cell broadening during cold-air
15 outbreaks. *Boundary-Layer Meteorology*, **81**, 289-323.
- 16 Muller, J., and Coauthors, 2008: Global isoprene emissions estimated using MEGAN, ECMWF analyses and
17 a detailed canopy environment model. *Atmospheric Chemistry and Physics*, **8**, 1329-1341.
- 18 Murphy, D. M., S. Solomon, R. W. Portmann, K. H. Rosenlof, P. M. Forster, and T. Wong, 2009: An
19 observationally based energy balance for the Earth since 1950. *Journal of Geophysical Research*, **114**,
20 D17107.
- 21 Murray, B. J., S. L. Broadley, T. W. Wilson, J. D. Atkinson, and R. H. Wills, 2011: Heterogeneous freezing
22 of water droplets containing kaolinite particles. *Atmospheric Chemistry and Physics*, **11**, 4191-4207.
- 23 Myhre, G., 2009: Consistency between satellite-derived and modeled estimates of the direct aerosol effect.
24 *Science*, **325**, 187-190.
- 25 Myhre, G., and Coauthors, 2007: Comparison of the radiative properties and direct radiative effect of
26 aerosols from a global aerosol model and remote sensing data over ocean. *Tellus B*, **59**, 115-129.
- 27 Myhre, G., and Coauthors, 2009: Modelled radiative forcing of the direct aerosol effect with multi-
28 observation evaluation. *Atmospheric Chemistry and Physics*, **9**, 1365-1392.
- 29 Nakajima, T., and S. Michael, 2009: What do we know about large scale changes of aerosols, clouds, and the
30 radiative budget? *Clouds in the Perturbed Climate System: Their Relationship to Energy Balance,*
31 *Atmospheric Dynamics, and Precipitation*, R. J. Charlson, and J. Heintzenberg, Eds., MIT Press.
- 32 Neale, R. B., J. H. Richter, and M. Jochum, 2008: The impact of convection on ENSO: From a delayed
33 oscillator to a series of events. *Journal of Climate*, **21**, 5904-5924.
- 34 Neelin, J. D., M. Münnich, H. Su, J. E. Meyerson, and C. E. Holloway, 2006: Tropical drying trends in
35 global warming models and observations. *Proc. Natl. Acad. Science of the USA*, **103**, 6110-6115.
- 36 Neggers, R. A. J., 2009: A dual mass flux framework for boundary layer convection. Part II: Clouds. *Journal*
37 *of the Atmospheric Sciences*, **66**, 1489-1506.
- 38 Neggers, R. A. J., M. Kohler, and A. C. M. Beljaars, 2009: A dual mass flux framework for boundary layer
39 convection. Part I: Transport. *Journal of the Atmospheric Sciences*, **66**, 1465-1487.
- 40 Nenes, A., and J. H. Seinfeld, 2003: Parameterization of cloud droplet formation in global climate models. *J.*
41 *Geophys. Res.*, **108**, 4415.
- 42 Nenes, A., S. N. Pandis, and C. Pilinis, 1998: ISORROPIA: A new thermodynamic equilibrium model for
43 multiphase multicomponent inorganic aerosols. *Aquatic Geochemistry*, **4**, 123-152.
- 44 Niedermeier, D., and Coauthors, 2010: Heterogeneous freezing of droplets with immersed mineral dust
45 particles - measurements and parameterization. *Atmospheric Chemistry and Physics*, **10**, 3601-3614.
- 46 Niemand, M., and Coauthors, 2011: Parameterization of immersion freezing on mineral dust particles: An
47 application in a regional scale model. *Journal of the Atmospheric Science*, *subm. to.*
- 48 Norris, J. R., and S. F. Iacobellis, 2005: North Pacific cloud feedbacks inferred from synoptic-scale dynamic
49 and thermodynamic relationships. *Journal of Climate*, **18**, 4862-4878.
- 50 Nuijens, L., B. Stevens, and A. P. Siebesma, 2009: The environment of precipitating shallow cumulus
51 convection. *Journal of the Atmospheric Sciences*, **66**, 1962-1979.
- 52 Nyanganyura, D., W. Maenhaut, M. Mathuthu, A. Makarau, and F. X. Meixner, 2007: The chemical
53 composition of tropospheric aerosols and their contributing sources to a continental background site in
54 northern Zimbabwe from 1994 to 2000. *Atmospheric Environment*, **41**, 2644-2659.
- 55 O'Dowd, C., C. Monahan, and M. Dall'Osto, 2010: On the occurrence of open ocean particle production and
56 growth events. *Geophysical Research Letters*, **37**, L19805.

- 1 O'Gorman, P., and T. Schneider, 2009: The physical basis for increases in precipitation extremes in
2 simulations of 21st-century climate change. *Proceedings of the National Academy of Sciences of the*
3 *United States of America*, **106**, 14773-14777.
- 4 Oanh, N. T. K., and Coauthors, 2006: Particulate air pollution in six Asian cities: Spatial and temporal
5 distributions, and associated sources. *Atmospheric Environment*, **40**, 3367-3380.
- 6 Oleson, K. W., G. B. Bonan, and J. Feddema, 2010: Effects of white roofs on urban temperature in a global
7 climate model. *Geophysical Research Letters*, **37**, L03701.
- 8 Omar, A. H., and Coauthors, 2009: The CALIPSO automated aerosol classification and lidar ratio selection
9 algorithm. *Journal of Atmospheric and Oceanic Technology*, **26**, 1994-2014.
- 10 Oouchi, K., A. T. Noda, M. Satoh, B. Wang, S. P. Xie, H. G. Takahashi, and T. Yasunari, 2009: Asian
11 summer monsoon simulated by a global cloud-system-resolving model: Diurnal to intra-seasonal
12 variability. *Geophysical Research Letters*, **36**, L11815.
- 13 Osborne, S. R., A. J. Barana, B. T. Johnson, J. M. Haywood, E. Hesse, and S. NeW man, 2011: Short-wave
14 and long-wave radiative properties of Saharan dust aerosol. *Quarterly Journal of the Royal*
15 *Meteorological Society*, **137**, 1149-1167.
- 16 Oshima, N., and Coauthors, 2011: Wet removal of black carbon in Asian outflow: Aerosol Radiative Forcing
17 in East Asia (A-FORCE) aircraft campaign. *Journal of Geophysical Research-Atmospheres*,
18 submitted.
- 19 Ovadnevaite, J., C. O'Dowd, M. Dall'Osto, D. Ceburnis, D. R. Worsnop, and H. Berresheim, 2011: Detecting
20 high contributions of primary organic matter to marine aerosol: A case study. *Geophysical Research*
21 *Letters*, **38**, L02807.
- 22 Paasonen, P., and Coauthors, 2010: On the roles of sulphuric acid and low-volatility organic vapours in the
23 initial steps of atmospheric new particle formation. *Atmospheric Chemistry and Physics*, **10**, 11223-
24 11242.
- 25 Pacifico, F., S. P. Harrison, C. D. Jones, and S. Sitch, 2009: Isoprene emissions and climate. *Atmospheric*
26 *Environment*, **43**, 6121-6135.
- 27 Painter, T. H., J. S. Deems, J. Belnap, A. F. Hamlet, C. C. Landry, and B. Udall, 2010: Response of
28 Colorado River runoff to dust radiative forcing in snow. *Proceedings of the National Academy of*
29 *Sciences of the United States of America*, **107**, 17125-17130.
- 30 Pallé, E., 2005: Possible satellite perspective effects on the reported correlations between solar activity and
31 clouds. *Geophysical Research Letters*, **32**, L03802.
- 32 Palm, S. P., S. T. Strey, J. Spinhirne, and T. Markus, 2010: Influence of Arctic sea ice extent on polar cloud
33 fraction and vertical structure and implications for regional climate. *Journal of Geophysical Research-*
34 *Atmospheres*, **115**, 9.
- 35 Park, S., and C. S. Bretherton, 2009: The University of Washington shallow convection and moist turbulence
36 schemes and their impact on climate simulations with the Community Atmosphere Model. *Journal of*
37 *Climate*, **22**, 3449-3469.
- 38 Pawlowska, H., and J. L. Brenguier, 2003: An observational study of drizzle formation in stratocumulus
39 clouds for general circulation model (GCM) parameterizations. *Journal of Geophysical Research*, **108**,
40 8630.
- 41 Pechony, O., and D. T. Shindell, 2010: Driving forces of global wildfires over the past millennium and the
42 forthcoming century. *Proceedings of the National Academy of Sciences of the United States of*
43 *America*, **107**, 19167-19170.
- 44 Peng, Y. R., and U. Lohmann, 2003: Sensitivity study of the spectral dispersion of the cloud droplet size
45 distribution on the indirect aerosol effect. *Geophysical Research Letters*, **30**, 1507.
- 46 Penner, J. E., S. Y. Zhang, and C. C. Chuang, 2003: Soot and smoke aerosol may not warm climate. *Journal*
47 *of Geophysical Research*, **108**, 4657.
- 48 Penner, J. E., L. Xu, and M. H. Wang, 2011: Satellite methods underestimate indirect climate forcing by
49 aerosols. *Proceedings of the National Academy of Sciences of the United States of America*, **108**,
50 13404-13408.
- 51 Penner, J. E., Y. Chen, M. Wang, and X. Liu, 2009: Possible influence of anthropogenic aerosols on cirrus
52 clouds and anthropogenic forcing. *Atmospheric Chemistry and Physics*, **9**, 879-896.
- 53 Penner, J. E., and Coauthors, 2006: Model intercomparison of indirect aerosol effects. *Atmospheric*
54 *Chemistry and Physics*, **6**, 3391-3405.
- 55 Penner, J. E., and Coauthors, 2001: Aerosols, their direct and indirect effects. *Climate Change 2001: The*
56 *Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the*
57 *Intergovernmental Panel on Climate Change*, Cambridge University Press.

- 1 Perez, N., J. Pey, X. Querol, A. Alastuey, J. M. Lopez, and M. Viana, 2008: Partitioning of major and trace
2 components in PM10-PM2.5-PM1 at an urban site in Southern Europe. *Atmospheric Environment*, **42**,
3 1677-1691.
- 4 Peters, K., J. Quaas, and N. Bellouin, 2011: Effects of absorbing aerosols in cloudy skies: a satellite study
5 over the Atlantic Ocean. *Atmospheric Chemistry and Physics*, **11**, 1393-1404.
- 6 Petroff, A., and L. Zhang, 2010: Development and validation of a size-resolved particle dry deposition
7 scheme for application in aerosol transport models. *Geoscientific Model Development*, **3**, 753-769.
- 8 Petters, M. D., and S. M. Kreidenweis, 2007: A single parameter representation of hygroscopic growth and
9 cloud condensation nucleus activity. *Atmospheric Chemistry and Physics*, **7**, 1961-1971.
- 10 Petters, M. D., J. R. Snider, B. Stevens, G. Vali, I. Faloona, and L. M. Russell, 2006: Accumulation mode
11 aerosol, pockets of open cells, and particle nucleation in the remote subtropical Pacific marine
12 boundary layer. *Journal of Geophysical Research-Atmospheres*, **111**.
- 13 Petters, M. D., and Coauthors, 2009: Ice nuclei emissions from biomass burning. *Journal of Geophysical
14 Research*, **114**, D07209.
- 15 Phillips, T. J., and Coauthors, 2004: Evaluating parameterizations in General Circulation Models: Climate
16 simulation meets weather prediction. *Bulletin of the American Meteorological Society*, **85**, 1903-1915.
- 17 Phillips, V. T. J., P. J. DeMott, and C. Andronache, 2008: An empirical parameterization of heterogeneous
18 ice nucleation for multiple chemical species of aerosol. *Journal of the Atmospheric Sciences*, **65**,
19 2757-2783.
- 20 Phillips, V. T. J., and Coauthors, 2009: Potential impacts from biological aerosols on ensembles of
21 continental clouds simulated numerically. *Biogeosciences*, **6**, 987-1014.
- 22 Pierce, J. R., and P. J. Adams, 2009a: Can cosmic rays affect cloud condensation nuclei by altering new
23 particle formation rates? *Geophysical Research Letters*, **36**, L09820.
- 24 —, 2009b: Uncertainty in global CCN concentrations from uncertain aerosol nucleation and primary
25 emission rates. *Atmospheric Chemistry and Physics*, **9**, 1339-1356.
- 26 Pierce, J. R., D. K. Weisenstein, P. Heckendorn, T. Peter, and D. W. Keith, 2010: Efficient formation of
27 stratospheric aerosol for climate engineering by emission of condensable vapor from aircraft.
28 *Geophysical Research Letters*, **37**, L18805.
- 29 Pincus, R., and M. B. Baker, 1994: Effect of precipitation on the albedo susceptibility of clouds in the marine
30 boundary-layer. *Nature*, **372**, 250-252.
- 31 Platnick, S., M. D. King, S. A. Ackerman, W. P. Menzel, B. A. Baum, J. C. Riedi, and R. A. Frey, 2003: The
32 MODIS cloud products: Algorithms and examples from Terra. *IEEE Transactions on Geoscience and
33 Remote Sensing*, **41**, 459-473.
- 34 Ponater, M., S. Marquart, R. Sausen, and U. Schumann, 2005: On contrail climate sensitivity. *Geophys. Res.
35 Lett.*, **32**, L10706.
- 36 Pósfai, M., R. Simonics, J. Li, P. Hobbs, and P. Buseck, 2003: Individual aerosol particles from biomass
37 burning in southern Africa: 1. Compositions and size distributions of carbonaceous particles. *Journal
38 of Geophysical Research-Atmospheres*, ARTN 8483, DOI 10.1029/2002JD002291.
- 39 Posselt, R., and U. Lohmann, 2008: Influence of giant CCN on warm rain processes in the ECHAM5 GCM.
40 *Atmospheric Chemistry and Physics*, **8**, 3769-3788.
- 41 Posselt, R., and U. Lohmann, 2009: Sensitivity of the total anthropogenic aerosol effect to the treatment of
42 rain in a global climate model. *Geophysical Research Letters*, **36**, L02805.
- 43 Pratt, K. A., and K. A. Prather, 2010: Aircraft measurements of vertical profiles of aerosol mixing states.
44 *Journal of Geophysical Research*, **115**, D11305.
- 45 Pratt, K. A., and Coauthors, 2009: In situ detection of biological particles in cloud ice-crystals. *Nature
46 Geoscience*, **2**, 397-400.
- 47 Prenni, A. J., and Coauthors, 2007: Can ice-nucleating aerosols affect arctic seasonal climate? *Bulletin of the
48 American Meteorological Society*, **88**, 541-+.
- 49 Prenni, A. J., and Coauthors, 2009: Relative roles of biogenic emissions and Saharan dust as ice nuclei in the
50 Amazon basin. *Nature Geoscience*, **2**, 401-404.
- 51 Pringle, K., H. Tost, A. Pozzer, U. Poschl, and J. Lelieveld, 2010: Global distribution of the effective aerosol
52 hygroscopicity parameter for CCN activation. *Atmospheric Chemistry and Physics*, 5241-5255.
- 53 Prisle, N. L., T. Raatikainen, A. Laaksonen, and M. Bilde, 2010: Surfactants in cloud droplet activation:
54 mixed organic-inorganic particles. *Atmospheric Chemistry and Physics*, **10**, 5663-5683.
- 55 Prisle, N. L., T. Raatikainen, R. Sorjamaa, B. Svenningsson, A. Laaksonen, and M. Bilde, 2008: Surfactant
56 partitioning in cloud droplet activation: a study of C8, C10, C12 and C14 normal fatty acid sodium
57 salts. *Tellus Series B-Chemical and Physical Meteorology*, **60**, 416-431.

- 1 Pritchard, M. S., and R. C. J. Somerville, 2010: Assessing the diurnal cycle of precipitation in a multi-scale
2 climate model. *J. Adv. Model. Earth Syst.*, **1**, Art. #12.
- 3 Puma, M. J., and B. I. Cook, 2010: Effects of irrigation on global climate during the 20th century. *J.*
4 *Geophys. Res.*, **115**, D16120.
- 5 Putaud, J.-P., and Coauthors, 2004: European aerosol phenomenology-2: chemical characteristics of
6 particulate matter at kerbside, urban, rural and background sites in Europe. *Atmospheric Environment*,
7 2579-2595.
- 8 Puxbaum, H., and Coauthors, 2004: A dual site study of PM_{2.5} and PM₁₀ aerosol chemistry in the larger
9 region of Vienna, Austria. *Atmos. Environ.*, **38**, 3949-3958.
- 10 Pye, H., H. Liao, S. Wu, L. Mickley, D. Jacob, D. Henze, and J. Seinfeld, 2009: Effect of changes in climate
11 and emissions on future sulfate-nitrate-ammonium aerosol levels in the United States. *Journal of*
12 *Geophysical Research-Atmospheres*, **114**, -.
- 13 Qu, W. J., X. Y. Zhang, R. Arimoto, D. Wang, Y. Q. Wang, L. W. Yan, and Y. Li, 2008: Chemical
14 composition of the background aerosol at two sites in southwestern and northwestern China: potential
15 influences of regional transport. *Tellus*, **60B**, 657-673.
- 16 Quaas, J., and O. Boucher, 2005: Constraining the first aerosol indirect radiative forcing in the LMDZ GCM
17 using POLDER and MODIS satellite data. *Geophysical Research Letters*, **32**, L17814.
- 18 Quaas, J., O. Boucher, and F. M. Bréon, 2004: Aerosol indirect effects in POLDER satellite data and the
19 Laboratoire de Meteorologie Dynamique-Zoom (LMDZ) general circulation model. *Journal of*
20 *Geophysical Research*, **109**, D08205.
- 21 Quaas, J., O. Boucher, and U. Lohmann, 2006: Constraining the total aerosol indirect effect in the LMDZ
22 and ECHAM4 GCMs using MODIS satellite data. *Atmospheric Chemistry and Physics*, **6**, 947-955.
- 23 Quaas, J., O. Boucher, N. Bellouin, and S. Kinne, 2008: Satellite-based estimate of the direct and indirect
24 aerosol climate forcing. *Journal of Geophysical Research-Atmospheres*, D05204, DOI
25 10.1029/2007JD008962.
- 26 Quaas, J., O. Boucher, A. Jones, G. Weedon, J. Kieser, and H. Joos, 2009a: Exploiting the weekly cycle as
27 observed over Europe to analyse aerosol indirect effects in two climate models. *Atmospheric*
28 *Chemistry and Physics*, **9**, 8493-8501.
- 29 Quaas, J., and Coauthors, 2009b: Aerosol indirect effects - general circulation model intercomparison and
30 evaluation with satellite data. *Atmospheric Chemistry and Physics*, **9**, 8697-8717.
- 31 Querol, X., and Coauthors, 2001: PM₁₀ and PM_{2.5} source apportionment in the Barcelona Metropolitan
32 Area, Catalonia, Spain. *Atmospheric Environment*, **35/36**, 6407-6419.
- 33 Querol, X., and Coauthors, 2006: Immobilization of heavy metals in polluted soils by the addition of zeolitic
34 material synthesized from coal fly ash. *Chemosphere*, **62**, 171-180.
- 35 Querol, X., and Coauthors, 2009: Variability in regional background aerosols within the Mediterranean.
36 *Atmos. Chem. Phys.*, **9**, 4575-4591.
- 37 Querol, X., and Coauthors, 2004: Speciation and origin of PM₁₀ and PM_{2.5} in selected European cities.
38 *Atmospheric Environment*, **38**, 6547-6555.
- 39 Querol, X., and Coauthors, 2008: Spatial and temporal variations in airborne particulate matter (PM₁₀ and
40 PM_{2.5}) across Spain 1999-2005. *Atmospheric Environment*.
- 41 Racherla, P., and P. Adams, 2006: Sensitivity of global tropospheric ozone and fine particulate matter
42 concentrations to climate change. *Journal of Geophysical Research-Atmospheres*, **111**, -.
- 43 Radhi, M., M. A. Box, G. P. Box, R. M. Mitchell, D. D. Cohen, E. Stelcer, and M. D. Keywood, 2010:
44 Optical, physical and chemical characteristics of Australian continental aerosols: results from a field
45 experiment. *Atmos. Chem. Phys.*, **10**, 5925-5942.
- 46 Rae, J. G. L., C. E. Johnson, N. Bellouin, O. Boucher, J. M. Haywood, and A. Jones, 2007: Sensitivity of
47 global sulphate aerosol production to changes in oxidant concentrations and climate. *Journal of*
48 *Geophysical Research*, **112**, D10312.
- 49 Ram, K., M. M. Sarin, and P. Hegde, 2010: Long-term record of aerosol optical properties and chemical
50 composition from a high-altitude site (Manora Peak) in Central Himalaya. *Atmos. Chem. Phys.*, **10**,
51 11791-11803.
- 52 Raman, R. S., S. Ramachandran, and N. Rastogi, 2010: Source identification of ambient aerosols over an
53 urban region in western India. *Journal of Environmental Monitoring*, **12**, 1330-1340.
- 54 Ramanathan, V., and G. Carmichael, 2008: Global and regional climate changes due to black carbon. *Nature*
55 *Geoscience*, **1**, 221-227.

- 1 Ramaswamy, V., and Coauthors, 2001: Radiative Forcing of Climate Change. *Climate Change 2001: The*
2 *Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the*
3 *Intergovernmental Panel on Climate Change*, Cambridge University Press.
- 4 Randall, D. A., 1989: Cloud parameterization for climate models: Status and prospects. *Atmospheric*
5 *Research*, **23**, 345-362.
- 6 Randall, D. A., and Coauthors, 2007: Climate Models and Their Evaluation. *Climate Change 2007: The*
7 *Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the*
8 *Intergovernmental Panel on Climate Change*, Cambridge University Press.
- 9 Rap, A., P. Forster, J. Haywood, A. Jones, and O. Boucher, 2010a: Estimating the climate impact of linear
10 contrails using the UK Met Office climate model. *Geophysical Research Letters*, ARTN L20703, DOI
11 10.1029/2010GL045161.
- 12 Rap, A., P. Forster, A. Jones, O. Boucher, J. Haywood, N. Bellouin, and R. De Leon, 2010b:
13 Parameterization of contrails in the UK Met Office Climate Model. *Journal of Geophysical Research-*
14 *Atmospheres*, **115**, D10205.
- 15 Rasch, P. J., P. J. Crutzen, and D. B. Coleman, 2008a: Exploring the geoengineering of climate using
16 stratospheric sulfate aerosols: The role of particle size. *Geophysical Research Letters*, **35**, L02809.
- 17 Rasch, P. J., C. C. Chen, and J. L. Latham, 2009: Geo-engineering by cloud seeding: influence on sea-ice
18 and the climate system. *Environmental Research Letters*, **4**, 045112.
- 19 Rasch, P. J., and Coauthors, 2008b: An overview of geoengineering of climate using stratospheric sulphate
20 aerosols. *Philosophical Transactions Royal Society London A*, **366**, 4007-4037.
- 21 Rastogi, N., and M. M. Sarin, 2005: Long-term characterization of ionic species in aerosols from urban and
22 high-altitude sites in western India: Role of mineral dust and anthropogenic sources. *Atmospheric*
23 *Environment*, **39**, 5541-5554.
- 24 Rauber, R. M., and Coauthors, 2007: Rain in shallow cumulus over the ocean - The RICO campaign.
25 *Bulletin of the American Meteorological Society*, **88**, 1912-1928.
- 26 Reddy, M. S., O. Boucher, Y. Balkanski, and M. Schulz, 2005: Aerosol optical depths and direct radiative
27 perturbations by species and source type. *Geophysical Research Letters*, **32**.
- 28 Remer, L. A., and Coauthors, 2005: The MODIS aerosol algorithm, products, and validation. *Journal of the*
29 *Atmospheric Sciences*, **62**, 947-973.
- 30 Rengarajan, R., M. M. Sarin, and A. K. Sudheer, 2007: Carbonaceous and inorganic species in atmospheric
31 aerosols during wintertime over urban and high-altitude sites in North India. *J. Geophysical Research*,
32 **112**.
- 33 Richter, J. H., and P. J. Rasch, 2008: Effects of convective momentum transport on the atmospheric
34 circulation in the community atmosphere model, version 3. *Journal of Climate*, **21**, 1487-1499.
- 35 Ridgwell, A., J. Singarayer, A. Hetherington, and P. Valdes, 2009: Tackling regional climate change by leaf
36 albedo bio-geoengineering. *Current Biology*, **19**, 146-150.
- 37 Riipinen, I., and Coauthors, 2011: Organic condensation: a vital link connecting aerosol formation to cloud
38 condensation nuclei (CCN) concentrations. *Atmospheric Chemistry and Physics*, **11**, 3865-3878.
- 39 Ringer, M. A., and Coauthors, 2006: Global mean cloud feedbacks in idealized climate change experiments.
40 *Geophysical Research Letters*, **33**, L07718.
- 41 Roberts, G. C., and Coauthors, 2010: Characterization of particle cloud droplet activity and composition in
42 the free troposphere and the boundary layer during INTEx-B. *Atmospheric Chemistry and Physics*,
43 **10**, 6627-6644.
- 44 Roberts, P., and J. Hallett, 1968: A LABORATORY STUDY OF ICE NUCLEATING PROPERTIES OF
45 SOME MINERAL PARTICULATES. *Quarterly Journal of the Royal Meteorological Society*, **94**, 25-
46 &.
- 47 Robinson, A., and Coauthors, 2007: Rethinking organic aerosols: Semivolatile emissions and photochemical
48 aging. *Science*, 1259-1262.
- 49 Robock, A., L. Oman, and G. Stenchikov, 2008: Regional climate responses to geoengineering with tropical
50 and Arctic SO₂ injections. *Journal of Geophysical Research*, **113**, D16101.
- 51 Rodriguez, S., X. Querol, A. Alastuey, and F. Plana, 2002: Sources and processes affecting levels and
52 composition of atmospheric aerosol in the western Mediterranean. *Journal of Geophysical Research*,
53 **107**, 4777.
- 54 Rodriguez, S., X. Querol, A. Alastuey, M. M. Viana, M. Alarcon, E. Mantilla, and C. R. Ruiz, 2004:
55 Comparative PM₁₀-PM_{2.5} source contribution study at rural, urban and industrial sites during PM
56 episodes in Eastern Spain. *Sci. Total Environ.*, **328**, 95-113.
- 57 Roe, J. H., and M. B. Baker, 2007: Why is climate sensitivity so unpredictable? *Science*, **318**, 629-632.

- 1 Rohs, S., R. Spang, F. Rohrer, C. Schiller, and H. Vos, 2010: A correlation study of high - altitude and
2 midaltitude clouds and galactic cosmic rays by MIPAS-Envisat. *J. Geophys. Res.*, **115**.
- 3 Romps, D. M., 2011: Response of tropical precipitation to global warming. *Journal of the Atmospheric*
4 *Sciences*, **68**, 123-138.
- 5 Rondanelli, R., and R. S. Lindzen, 2010: Can thin cirrus clouds in the tropics provide a solution to the faint
6 young Sun paradox? *Journal of Geophysical Research-Atmospheres*, **115**, 12.
- 7 Roosli, M., and Coauthors, 2001: Temporal and spatial variation of the chemical composition of PM10 at
8 urban and rural sites in the Basel area, Switzerland. *Atmospheric Environment*, **35**, 3701-3713.
- 9 Rose, D., and Coauthors, 2011: Cloud condensation nuclei in polluted air and biomass burning smoke near
10 the mega-city Guangzhou, China -Part 2: Size-resolved aerosol chemical composition, diurnal cycles,
11 and externally mixed weakly CCN-active soot particles. *Atmospheric Chemistry and Physics*, **11**,
12 2817-2836.
- 13 Rosenfeld, A., H. Akbari, J. Romm, and M. Pomerantz, 1998: Cool communities: strategies for heat island
14 mitigation and smog reduction. *Energy and Buildings*, **28**, 51-62.
- 15 Rosenfeld, D., and W. L. Woodley, 2001: Pollution and clouds. *Physics World*, **14**, 33-37.
- 16 Rosenfeld, D., Y. J. Kaufman, and I. Koren, 2006: Switching cloud cover and dynamical regimes from open
17 to closed Benard cells in response to the suppression of precipitation by aerosols. *Atmospheric*
18 *Chemistry and Physics*, **6**, 2503-2511.
- 19 Rosenfeld, D., and Coauthors, 2008: Flood or drought: How do aerosols affect precipitation? *Science*, **321**,
20 1309-1313.
- 21 Rotstayn, L. D., 1999: Indirect forcing by anthropogenic aerosols: A global climate model calculation of the
22 effective-radius and cloud-lifetime effects. *Journal of Geophysical Research-Atmospheres*, **104**, 9369-
23 9380.
- 24 Rotstayn, L. D., and J. E. Penner, 2001: Indirect aerosol forcing, quasi forcing, and climate response. *Journal*
25 *of Climate*, **14**, 2960-2975.
- 26 Rotstayn, L. D., and Y. G. Liu, 2003: Sensitivity of the first indirect aerosol effect to an increase of cloud
27 droplet spectral dispersion with droplet number concentration. *Journal of Climate*, **16**, 3476-3481.
- 28 ———, 2005: A smaller global estimate of the second indirect aerosol effect. *Geophysical Research Letters*,
29 **32**.
- 30 ———, 2009: Cloud droplet spectral dispersion and the indirect aerosol effect: Comparison of two treatments
31 in a GCM. *Geophysical Research Letters*, **36**.
- 32 Rotstayn, L. D., and Coauthors, 2007: Have Australian rainfall and cloudiness increased due to the remote
33 effects of Asian anthropogenic aerosols? *Journal of Geophysical Research-Atmospheres*, **112**.
- 34 Russell, M., and D. Allen, 2005: Predicting secondary organic aerosol formation rates in southeast Texas.
35 *Journal of Geophysical Research-Atmospheres*, ARTN D07S17, DOI 10.1029/2004JD004722.
- 36 Rypdal, K., N. Rive, T. K. Berntsen, Z. Klimont, T. K. Mideksa, G. Myhre, and R. B. Skeie, 2009: Costs and
37 global impacts of black carbon abatement strategies. *Tellus*, **61B**, 625-641.
- 38 Sacks, W. J., B. I. Cook, N. Buening, S. Levis, and J. H. Helkowski, 2009: Effects of global irrigation on
39 the near-surface climate. *Climate Dynamics*, **33**, 159-175.
- 40 Safai, P. D., K. B. Budhavant, P. S. P. Rao, K. Ali, and A. Sinha, 2010: Source characterization for aerosol
41 constituents and changing roles of calcium and ammonium aerosols in the neutralization of aerosol
42 acidity at a semi-urban site in SW India. *Atmospheric Research*, **98**, 78-88.
- 43 Sakaeda, N., R. Wood, and P. Rasch, 2011: Direct and semidirect aerosol effects of southern African
44 biomass burning aerosol. *Journal of Geophysical Research-Atmospheres*, **116**.
- 45 Salam, A., U. Lohmann, and G. Lesins, 2007: Ice nucleation of ammonia gas exposed montmorillonite
46 mineral dust particles. *Atmospheric Chemistry and Physics*, **7**, 3923-3931.
- 47 Salam, A., and Coauthors, 2006: Ice nucleation studies of mineral dust particles with a new continuous flow
48 diffusion chamber. *Aerosol Science and Technology*, **40**, 134-143.
- 49 Salvador, P., B. Artinano, X. Querol, A. Alastuey, and M. Costoya, 2007: Characterisation of local and
50 external contributions of atmospheric particulate matter at a background coastal site. *Atmospheric*
51 *Environment*, **41**, 1-17.
- 52 Salzmann, M., and Coauthors, 2010: Two-moment bulk stratiform cloud microphysics in the GFDL AM3
53 GCM: description, evaluation, and sensitivity tests. *Atmospheric Chemistry and Physics*, 8037-8064.
- 54 Sanchez-Lorenzo, A., J. Calbó, J. Martin - Vide, A. Garcia - Manuel, G. Garcia - Soriano, and C. Beck,
55 2008: Winter “weekend effect” in southern Europe and its connections with periodicities in
56 atmospheric dynamics. *Geophys. Res. Lett.*, **35**, L15711.

- 1 Sanderson, M., C. Jones, W. Collins, C. Johnson, and R. Derwent, 2003: Effect of climate change on
2 isoprene emissions and surface ozone levels. *Geophysical Research Letters*, **30**, -.
- 3 Sassen, K., and G. C. Dodd, 1988: Homogeneous nucleation rate for highly supercooled cirrus cloud
4 droplets. *Journal of the Atmospheric Sciences*, **45**, 1357-1369.
- 5 Sassen, K., P. J. DeMott, J. M. Prospero, and M. R. Poellot, 2003: Saharan dust storms and indirect aerosol
6 effects on clouds: CRYSTAL-FACE results. *Geophysical Research Letters*, **30**.
- 7 Satheesh, S. K., and K. K. Moorthy, 2005: Radiative effects of natural aerosols: A review. *Atmospheric*
8 *Environment*, **39**, 2089-2110.
- 9 Sato, T., H. Miura, M. Satoh, Y. Takayabu, and Y. Wang, 2009: Diurnal cycle of precipitation in the Tropics
10 simulated in a global cloud-resolving model. *Journal of Climate*, **22**, 4809-4826.
- 11 Savic-Jovicic, V., and B. Stevens, 2008: The structure and mesoscale organization of precipitating
12 stratocumulus. *Journal of the Atmospheric Sciences*, **65**, 1587-1605.
- 13 Sawant, A. A., K. Na, X. Zhu, and D. R. Cocker III, 2004: Chemical characterization of outdoor PM2.5 and
14 gas-phase compounds in Mira Loma, California. *Atmospheric Environment*, **38**, 5517-5528.
- 15 Schaller, R. C., and N. Fukuta, 1979: Ice nucleation by aerosol particles - Experimental studies using a
16 wedge-shaped ice thermal-diffusion chamber. *Journal of the Atmospheric Sciences*, **36**, 1788-1802.
- 17 Scheff, J., and D. Frierson, 2011: 21st-century multi-model subtropical precipitation declines are mostly
18 mid-latitude shifts. *Journal of Climate*, Submitted.
- 19 Schmidt, K. S., G. Feingold, P. Pilewskie, H. Jiang, O. Coddington, and M. Wendisch, 2009: Irradiance in
20 polluted cumulus fields: Measured and modeled cloud-aerosol effects. *Geophysical Research Letters*,
21 **36**.
- 22 Schreier, M., H. Mannstein, V. Eyring, and H. Bovensmann, 2007: Global ship track distribution and
23 radiative forcing from 1 year of AATSR data. *Geophysical Research Letters*, **34**.
- 24 Schwarz, J., and Coauthors, 2008: Coatings and their enhancement of black carbon light absorption in the
25 tropical atmosphere. *Journal of Geophysical Research-Atmospheres*, ARTN D03203, DOI
26 10.1029/2007JD009042.
- 27 Schwarz, J. P., and Coauthors, 2010: Global-scale black carbon profiles observed in the remote atmosphere
28 and compared to models. *Geophysical Research Letters*, **37**, L18812.
- 29 Schwarz, J. P., and Coauthors, 2006: Single-particle measurements of midlatitude black carbon and light-
30 scattering aerosols from the boundary layer to the lower stratosphere. *Journal of Geophysical*
31 *Research*, **111**, D16207.
- 32 Seifert, A., and B. Stevens, 2010: Microphysical scaling relations in a kinematic model of isolated shallow
33 cumulus clouds. *Journal of the Atmospheric Sciences*, **67**, 1575-1590.
- 34 Seifert, A., C. Köhler, and K. D. Beheng, 2011: Aerosol-cloud-precipitation effects over Germany as
35 simulated by a convective-scale numerical weather prediction model. *Atmos. Chem. Phys. Discuss.*,
36 **11**, 20203-20243.
- 37 Seifert, P., and Coauthors, 2010: Saharan dust and heterogeneous ice formation: Eleven years of cloud
38 observations at a central European EARLINET site. *Journal of Geophysical Research-Atmospheres*,
39 **115**.
- 40 Seitz, R., 2011: Bright water: hydrosols, water conservation and climate change. *Climatic Change*, **105**, 365-
41 381.
- 42 Sekiguchi, M., and Coauthors, 2003: A study of the direct and indirect effects of aerosols using global
43 satellite data sets of aerosol and cloud parameters. *Journal of Geophysical Research-Atmospheres*,
44 **108**.
- 45 Senior, C. A., and J. F. B. Mitchell, 1993: Carbon dioxide and climate: The impact of cloud
46 parameterization. *Journal of Climate*, **6**, 393-418.
- 47 ———, 2000: The time-dependence of climate sensitivity. *Geophysical Research Letters*, **27**, 2685-2688.
- 48 Sesartic, A., U. Lohmann, and T. Storelvmo, 2011: Bacteria in the ECHAM5-HAM global climate model.
49 *Atmospheric Chemistry Physics Discussion*, **11**, 1457-1488.
- 50 Sharon, T. M., and Coauthors, 2006: Aerosol and cloud microphysical characteristics of rifts and gradients in
51 maritime stratocumulus clouds. *Journal of the Atmospheric Sciences*, **63**, 983-997.
- 52 Shaw, R. A., A. J. Durant, and Y. Mi, 2005: Heterogeneous surface crystallization observed in undercooled
53 water. *Journal of Physical Chemistry B*, **109**, 9865-9868.
- 54 Shaw, S. B., A. A. Royem, and S. J. Riha, 2011: The relationship between extreme hourly precipitation and
55 surface temperature in different hydroclimatic regions of the United States. *Journal of*
56 *Hydrometeorology*, **12**, 319-325.

- 1 Sherwood, S. C., W. Ingram, Y. Tsushima, M. Satoh, M. Roberts, P. L. Vidale, and P. A. O’Gorman, 2010:
2 Relative humidity changes in a warmer climate. *Journal of Geophysical Research*, **115**, D09104.
- 3 Shilling, J. E., T. J. Fortin, and M. A. Tolbert, 2006: Depositional ice nucleation on crystalline organic and
4 inorganic solids. *Journal of Geophysical Research-Atmospheres*, **111**.
- 5 Shindell, D., and G. Faluvegi, 2009: Climate response to regional radiative forcing during the twentieth
6 century. *Nature Geoscience*, **2**, 294-300.
- 7 Shiraiwa, M., and Coauthors, 2008: Radiative impact of mixing state of black carbon aerosol in Asian
8 outflow. *Journal of Geophysical Research*, **113**, D24210.
- 9 Shresth, A. B., C. P. Wake, J. E. Dibb, P. A. Mayewski, S. I. Whitlow, G. R. Carmichael, and M. Ferm,
10 2000: Seasonal variations in aerosol concentrations and compositions in the Nepal Himalaya.
11 *Atmospheric Environment*, **34**, 3349-3363.
- 12 Shupe, M. D., and Coauthors, 2008: A focus on mixed-phase clouds. The status of ground-based
13 observational methods. *Bulletin of the American Meteorological Society*, **89**, 1549-1562.
- 14 Siebesma, A. P., P. M. M. Soares, and J. Teixeira, 2007: A combined eddy-diffusivity mass-flux approach
15 for the convective boundary layer. *Journal of the Atmospheric Sciences*, **64**, 1230-1248.
- 16 Siebesma, A. P., and Coauthors, 2009: Cloud-controlling factors. *Clouds in the Perturbed Climate System:
17 Their Relationship to Energy Balance, Atmospheric Dynamics, and Precipitation*, J. Heintzenberg, and
18 R. J. Charlson, Eds., MIT Press, 269-290.
- 19 Singh, H. B., W. H. Brune, J. H. Crawford, D. J. Jacob, and P. B. Russell, 2006: Overview of the summer
20 2004 Intercontinental Chemical Transport Experiment–North America (INTEX-A). *J. Geophys. Res.*,
21 **111**.
- 22 Sipila, M., and Coauthors, 2010: The role of sulfuric acid in atmospheric nucleation. *Science*, **327**, 1243-
23 1246.
- 24 Slingo, A., 1990: Sensitivity of the Earth’s radiation budget to changes in low clouds. *Nature*, **343**, 49-51.
- 25 Small, J. D., P. Y. Chuang, G. Feingold, and H. Jiang, 2009: Can aerosol decrease cloud lifetime?
26 *Geophysical Research Letters*, **36**.
- 27 Smirnov, A., and Coauthors, 2011: Maritime aerosol network as a component of AERONET - first results
28 and comparison with global aerosol models and satellite retrievals. *Atmospheric Measurement
29 Techniques*, **4**, 583-597.
- 30 Smith, J., and Coauthors, 2010: Observations of aminium salts in atmospheric nanoparticles and possible
31 climatic implications. *Proceedings of the National Academy of Sciences of the United States of
32 America*, **107**, 6634-6639.
- 33 Snow-Kroppla, E., J. Pierce, D. Westervelt, and W. Trivitayanurak, 2011: Cosmic rays, aerosol formation and
34 cloud-condensation nuclei: sensitivities to model uncertainties. *Atmospheric Chemistry and Physics*,
35 **11**, 4001-4013.
- 36 Soden, B. J., and I. M. Held, 2006: An assessment of climate feedbacks in coupled ocean-atmosphere
37 models. *Journal of Climate*, **19**, 3354-3360.
- 38 Soden, B. J., and G. A. Vecchi, 2011: The vertical distribution of cloud feedback in coupled ocean-
39 atmosphere models. *Geophysical Research Letters*, **38**, L12704.
- 40 Song, X., and G. J. Zhang, 2011: Microphysics parameterization for convective clouds in a global climate
41 model: Description and single-column model tests. *Journal of Geophysical Research*, **116**, D02201.
- 42 Souza, P. A. d., W. Z. d. Mello, L. MarianiRauda, and M. SellaSilvia, 2010: Characterization of fine and
43 coarse particulate matter and composition of the water-soluble inorganic fraction in São José dos
44 Campos (SP). *Quím. Nova*, **33**.
- 45 Spencer, R. W., and W. D. Braswell, 2008: Potential Biases in Feedback Diagnosis from Observational Data:
46 A Simple Model Demonstration. *Journal of Climate*, **21**, 5624-5628.
- 47 Spencer, R. W., and W. D. Braswell, 2010: On the diagnosis of radiative feedback in the presence of
48 unknown radiative forcing. *Journal of Geophysical Research-Atmospheres*, **115**.
- 49 Spracklen, D., K. Pringle, K. Carslaw, M. Chipperfield, and G. Mann, 2005a: A global off-line model of
50 size-resolved aerosol microphysics: II. Identification of key uncertainties. *Atmospheric Chemistry and
51 Physics*, 3233-3250.
- 52 —, 2005b: A global off-line model of size-resolved aerosol microphysics: I. Model development and
53 prediction of aerosol properties. *Atmospheric Chemistry and Physics*, 2227-2252.
- 54 Spracklen, D., K. Carslaw, M. Kulmala, V. Kerminen, G. Mann, and S. Sihto, 2006: The contribution of
55 boundary layer nucleation events to total particle concentrations on regional and global scales.
56 *Atmospheric Chemistry and Physics*, **6**, 5631-5648.

- 1 Spracklen, D., and Coauthors, 2008: Contribution of particle formation to global cloud condensation nuclei
2 concentrations. *Geophysical Research Letters*, ARTN L06808, DOI 10.1029/2007GL033038.
- 3 Spracklen, D., and Coauthors, 2010: Explaining global surface aerosol number concentrations in terms of
4 primary emissions and particle formation. *Atmospheric Chemistry and Physics*, 4775-4793.
- 5 Spracklen, D. V., and Coauthors, 2011: Aerosol mass spectrometer constraint on the global secondary
6 organic aerosol budget. *Atmos. Chem. Phys. Discuss.*, **11**, 5699-5755.
- 7 Stan, C., and Coauthors, 2010: An ocean-atmosphere climate simulation with an embedded cloud resolving
8 model. *Geophysical Research Letters*, **37**, L01702.
- 9 Stephens, G., and T. Ellis, 2008: Controls of global-mean precipitation increases in global warming GCM
10 experiments. *Journal of Climate*, **21**, 6141-6155.
- 11 Stephens, G. L., and C. D. Kummerow, 2007: The remote sensing of clouds and precipitation from space: A
12 review. *Journal of the Atmospheric Sciences*, **64**, 3742-3765.
- 13 Stephens, G. L., and Coauthors, 2002: The Cloudsat mission and the A-train - A new dimension of space-
14 based observations of clouds and precipitation. *Bulletin of the American Meteorological Society*, **83**,
15 1771-1790.
- 16 Stevens, B., and A. Seifert, 2008: Understanding macrophysical outcomes of microphysical choices in
17 simulations of shallow cumulus convection. *Journal of the Meteorological Society of Japan*, **86**, 143-
18 162.
- 19 Stevens, B., and G. Feingold, 2009: Untangling aerosol effects on clouds and precipitation in a buffered
20 system. *Nature*, **461**, 607-613.
- 21 Stevens, B., and J.-L. Brenguier, 2009: Cloud-controlling factors: Low clouds. *Clouds in the Perturbed
22 Climate System: Their Relationship to Energy Balance, Atmospheric Dynamics, and Precipitation*, J.
23 Heintzenberg, and R. J. Charlson, Eds., MIT Press, 173-196.
- 24 Stevens, B., W. R. Cotton, G. Feingold, and C.-H. Moeng, 1998: Large-eddy simulations of strongly
25 precipitating, shallow, stratocumulus-topped boundary layers. *Journal of the Atmospheric Sciences*,
26 **55**, 3616-3638.
- 27 Stevens, B., and Coauthors, 2005: Pockets of open cells and drizzle in marine stratocumulus. *Bulletin of the
28 American Meteorological Society*, **86**, 51-57.
- 29 Stier, P., J. Feichter, S. Kloster, E. Vignati, and J. Wilson, 2006a: Emission-induced nonlinearities in the
30 global aerosol system: Results from the ECHAM5-HAM aerosol-climate model. *Journal of Climate*,
31 **19**, 3845-3862.
- 32 Stier, P., J. H. Seinfeld, S. Kinne, J. Feichter, and O. Boucher, 2006b: Impact of nonabsorbing anthropogenic
33 aerosols on clear-sky atmospheric absorption. *Journal of Geophysical Research*, **111**, D18201.
- 34 Stier, P., and Coauthors, 2005: The aerosol-climate model ECHAM5-HAM. *Atmospheric Chemistry and
35 Physics*, 1125-1156.
- 36 Stjern, C. W., 2011: Weekly cycles in precipitation and other meteorological variables in a polluted region of
37 Europe. *Atmos. Chem. Phys.*, **11**, 4095-4104.
- 38 Stone, E., J. Schauer, T. A. Quraishi, and A. Mahmood, 2010: Chemical characterization and source
39 apportionment of fine and coarse particulate matter in Lahore, Pakistan. *Atmospheric Environment*, **44**,
40 1062-1070.
- 41 Stone, R. S., and Coauthors, 2008: Radiative impact of boreal smoke in the Arctic: Observed and modeled.
42 *Journal of Geophysical Research-Atmospheres*, **113**.
- 43 Storelvmo, T., 2011: Uncertainties in aerosol direct and indirect effects attributed to uncertainties in
44 convective transport parametrizations. *Atmospheric Research*, **in press**.
- 45 Storelvmo, T., J. E. Kristjansson, and U. Lohmann, 2008a: Aerosol influence on mixed-phase clouds in
46 CAM-Oslo. *Journal of the Atmospheric Sciences*, **65**, 3214-3230.
- 47 Storelvmo, T., U. Lohmann, and R. Bennartz, 2009: What governs the spread in shortwave forcings in the
48 transient IPCC AR4 models? *Geophysical Research Letters*, **36**, L01806.
- 49 Storelvmo, T., C. Hoose, and P. Eriksson, 2011: Global modeling of mixed-phase clouds: The albedo and
50 lifetime effects of aerosols. *Journal of Geophysical Research-Atmospheres*, **116**.
- 51 Storelvmo, T., J. E. Kristjansson, S. J. Ghan, A. Kirkevåg, O. Seland, and T. Iversen, 2006: Predicting cloud
52 droplet number concentration in Community Atmosphere Model (CAM)-Oslo. *Journal of Geophysical
53 Research-Atmospheres*, **111**.
- 54 Storelvmo, T., J. E. Kristjansson, U. Lohmann, T. Iversen, A. Kirkevåg, and O. Seland, 2008b: Modeling of
55 the Wegener-Bergeron-Findeisen process-implications for aerosol indirect effects. *Environmental
56 Research Letters*, **3**, 045001.

- 1 Stott, P. A., J. F. B. Mitchell, M. R. Allen, T. L. Delworth, J. M. Gregory, G. A. Meehl, and B. D. Santer,
2 2006: Observational constraints on past attributable warming and predictions of future global
3 warming. *Journal of Climate*, **19**, 3055-3069.
- 4 Stramler, K., A. D. Del Genio, and W. B. Rossow, 2011: Synoptically driven Arctic winter states. *Journal of*
5 *Climate*, **24**, 1747-1762.
- 6 Stratmann, F., O. Moehler, R. Shaw, and W. Heike, 2009: Laboratory cloud simulation: Capabilities and
7 future directions. *Clouds in the Perturbed Climate System: Their Relationship to Energy Balance,*
8 *Atmospheric Dynamics, and Precipitation*, J. Heintzenberg, and R. J. Charlson, Eds., MIT Press, 149-
9 172.
- 10 Stuber, N., and P. Forster, 2007: The impact of diurnal variations of air traffic on contrail radiative forcing.
11 *Atmospheric Chemistry and Physics*, 3153-3162.
- 12 Sullivan, R. C., L. Minambres, P. J. DeMott, A. J. Prenni, C. M. Carrico, E. J. T. Levin, and S. M.
13 Kreidenweis, 2010a: Chemical processing does not always impair heterogeneous ice nucleation of
14 mineral dust particles. *Geophysical Research Letters*, **37**, L24805.
- 15 Sullivan, R. C., and Coauthors, 2010b: Irreversible loss of ice nucleation active sites in mineral dust particles
16 caused by sulphuric acid condensation. *Atmospheric Chemistry and Physics*, **10**, 11471-11487.
- 17 Sun, D. L., K. M. Lau, and M. Kafatos, 2008: Contrasting the 2007 and 2005 hurricane seasons: Evidence of
18 possible impacts of Saharan dry air and dust on tropical cyclone activity in the Atlantic basin.
19 *Geophysical Research Letters*, **35**, L15405.
- 20 Sun, J. M., P. A. Ariya, H. G. Leighton, and M. K. Yau, 2010: Mystery of ice multiplication in warm-based
21 precipitating shallow cumulus clouds. *Geophysical Research Letters*, **37**, L10802.
- 22 Suzuki, K., T. Nakajima, A. Numaguti, T. Takemura, K. Kawamoto, and A. Higurashi, 2004: A study of the
23 aerosol effect on a cloud field with simultaneous use of GCM modeling and satellite observation.
24 *Journal of the Atmospheric Sciences*, **61**, 179-194.
- 25 Svensmark, H., and E. FriisChristensen, 1997: Variation of cosmic ray flux and global cloud coverage - A
26 missing link in solar-climate relationships. *Journal of Atmospheric and Solar-Terrestrial Physics*,
27 1225-1232.
- 28 Svensmark, H., T. Bondo, and J. Svensmark, 2009: Cosmic ray decreases affect atmospheric aerosols and
29 clouds. *Geophysical Research Letters*, ARTN L15101, DOI 10.1029/2009GL038429.
- 30 Takahashi, K., 2009: The global hydrological cycle and atmospheric shortwave absorption in climate models
31 under CO₂ forcing. *Journal of Climate*, **22**, 5667-5675.
- 32 Takemura, T., T. Nozawa, S. Emori, T. Y. Nakajima, and T. Nakajima, 2005: Simulation of climate response
33 to aerosol direct and indirect effects with aerosol transport-radiation model. *Journal of Geophysical*
34 *Research*, **110**, D02202.
- 35 Tanré, D., M. Herman, and Y. J. Kaufman, 1996: Information on aerosol size distribution contained in solar
36 reflected spectral radiances. *Journal of Geophysical Research-Atmospheres*, **101**, 19043-19060.
- 37 Tanré, D., Y. Kaufman, M. Herman, and S. Mattoo, 1997: Remote sensing of aerosol properties over oceans
38 using the MODIS/EOS spectral radiances. *Journal of Geophysical Research-Atmospheres*, 16971-
39 16988.
- 40 Tanré, D., and Coauthors, 2011: Remote sensing of aerosols by using polarized, directional and spectral
41 measurements within the A-Train: the PARASOL mission. *Atmospheric Measurement Techniques*, **4**,
42 1383-1395.
- 43 Tao, W.-K., R. J. Houze, and E. Smith, 2007: The fourth TRMM Latent Heating Workshop. *Bull. Amer.*
44 *Meteor. Soc.*, **88**, 1255-1259.
- 45 Tao, W.-K., and Coauthors, 2009: A Multiscale Modeling System: Developments, applications, and critical
46 issues. *Bull. Amer. Meteor. Soc.*, **90**, 515-534.
- 47 Targino, A. C., and Coauthors, 2009: Influence of particle chemical composition on the phase of cold clouds
48 at a high-alpine site in Switzerland. *Journal of Geophysical Research-Atmospheres*, **114**.
- 49 Tegen, I., M. Werner, S. Harrison, and K. Kohfeld, 2004: Relative importance of climate and land use in
50 determining present and future global soil dust emission. *Geophysical Research Letters*, **31**, -.
- 51 Theodosi, C., U. Im, A. Bougiatioti, P. Zarmas, O. Yenigun, and N. Mihalopoulos, 2010: Aerosol chemical
52 composition over Istanbul. *Science of the Total Environment*, **408**, 2482-2491.
- 53 Thomas, M. A., and Coauthors, 2010: Quantification of DMS aerosol-cloud-climate interactions using the
54 ECHAM5-HAMMOZ model in a current climate scenario. *Atmospheric Chemistry and Physics*, **10**,
55 7425-7438.
- 56 Tilmes, S., R. R. Garcia, E. D. Kinnison, A. Gettelman, and P. J. Rasch, 2009: Impact of geo-engineered
57 aerosols on troposphere and stratosphere. *Journal of Geophysical Research*, **114**, D12305.

- 1 Tomita, H., H. Miura, S. Iga, T. Nasuno, and M. Satoh, 2005: A global cloud-resolving simulation:
2 Preliminary results from an aqua planet experiment. *Geophysical Research Letters*, -.
- 3 Tompkins, A. M., K. Gierens, and G. Radel, 2007: Ice supersaturation in the ECMWF integrated forecast
4 system. *Quarterly Journal of the Royal Meteorological Society*, **133**, 53-63.
- 5 Torres, O., P. K. Bhartia, J. R. Herman, Z. Ahmad, and J. Gleason, 1998: Derivation of aerosol properties
6 from satellite measurements of backscattered ultraviolet radiation: Theoretical basis. *Journal of*
7 *Geophysical Research*, **103**, 17099-17110.
- 8 Torres, O., P. K. Bhartia, J. R. Herman, A. Sinyuk, P. Ginoux, and B. Holben, 2002: A long-term record of
9 aerosol optical depth from TOMS observations and comparison to AERONET measurements. *Journal*
10 *of the Atmospheric Sciences*, **59**, 398-413.
- 11 Trenberth, K. E., and A. Dai, 2007: Effects of Mount Pinatubo volcanic eruption on the hydrological cycle as
12 an analog of geoengineering. *Geophysical Research Letters*, **34**, L15702.
- 13 Trenberth, K. E., and J. T. Fasullo, 2009: Global warming due to increasing absorbed solar radiation.
14 *Geophysical Research Letters*, **36**, L07706.
- 15 ———, 2010: Simulation of present-day and twenty-first-century energy budgets of the Southern Oceans.
16 *Journal of Climate*, **23**, 440-454.
- 17 Tselioudis, G., and W. B. Rossow, 2006: Climate feedback implied by observed radiation and precipitation
18 changes with midlatitude storm strength and frequency. *Geophysical Research Letters*, **33**, 5.
- 19 Tsigaridis, K., and M. Kanakidou, 2003: Global modelling of secondary organic aerosol in the troposphere: a
20 sensitivity analysis. *Atmospheric Chemistry and Physics*, 1849-1869.
- 21 ———, 2007: Secondary organic aerosol importance in the future atmosphere. *Atmospheric Environment*,
22 4682-4692.
- 23 Tsushima, Y., and Coauthors, 2006: Importance of the mixed-phase cloud distribution in the control
24 climate for assessing the response of clouds to carbon dioxide increase: a multi-model study. *Climate*
25 *Dynamics*, **27**, 113-126.
- 26 Twohy, C. H., J. A. Coakley, Jr., and W. R. Tahnk, 2009: Effect of changes in relative humidity on aerosol
27 scattering near clouds. *Journal of Geophysical Research-Atmospheres*, **114**.
- 28 Twohy, C. H., and Coauthors, 2005: Evaluation of the aerosol indirect effect in marine stratocumulus clouds:
29 Droplet number, size, liquid water path, and radiative impact. *J. Geophys. Res.*, **110**, D08203.
- 30 Twohy, C. H., and Coauthors, 2010: Relationships of biomass-burning aerosols to ice in orographic wave
31 clouds. *Journal of the Atmospheric Sciences*, **67**, 2437-2450.
- 32 Twomey, S., 1977: Influence of pollution on shortwave albedo of clouds. *Journal of the Atmospheric*
33 *Sciences*, **34**, 1149-1152.
- 34 Udelhofen, P., and R. Cess, 2001: Cloud cover variations over the United States: An influence of cosmic
35 rays or solar variability? *Geophysical Research Letters*, **28**, 2617-2620.
- 36 Ulbrich, I., M. Canagaratna, Q. Zhang, D. Worsnop, and J. Jimenez, 2009: Interpretation of organic
37 components from Positive Matrix Factorization of aerosol mass spectrometric data. *Atmospheric*
38 *Chemistry and Physics*, 2891-2918.
- 39 Unger, N., S. Menon, D. M. Koch, and D. T. Shindell, 2009: Impacts of aerosol-cloud interactions on past
40 and future changes in tropospheric composition. *Atmospheric Chemistry and Physics*, **9**, 4115-4129.
- 41 Unger, N., D. Shindell, D. Koch, M. Amann, J. Cofala, and D. Streets, 2006: Influences of man-made
42 emissions and climate changes on tropospheric ozone, methane, and sulfate at 2030 from a broad
43 range of possible futures. *Journal of Geophysical Research-Atmospheres*, **111**, -.
- 44 Usoskin, I., and G. Kovaltsov, 2008: Cosmic rays and climate of the Earth: Possible connection. *Comptes*
45 *Rendus Geoscience*, **340**, 441-450.
- 46 Vali, G., 2008: Repeatability and randomness in heterogeneous freezing nucleation. *Atmospheric Chemistry*
47 *and Physics*, **8**, 5017-5031.
- 48 vanZanten, M. C., B. Stevens, G. Vali, and D. H. Lenschow, 2005: Observations of drizzle in nocturnal
49 marine stratocumulus. *Journal of the Atmospheric Sciences*, **62**, 88-106.
- 50 vanZanten, M. C., and Coauthors, 2011: Controls on precipitation and cloudiness in simulations of trade-
51 wind cumulus as observed during RICO. *Journal of Advances in Modeling Earth Systems*, **3**, 13 pp.
- 52 Várnai, T., and A. Marshak, 2009: MODIS observations of enhanced clear sky reflectance near clouds.
53 *Geophysical Research Letters*, **36**.
- 54 Vavrus, S., M. M. Holland, and D. A. Bailey, 2011: Changes in Arctic clouds during intervals of rapid sea
55 ice loss. *Climate Dynamics*, **36**, 1475-1489.

- 1 Vavrus, S., D. Waliser, A. Schweiger, and J. Francis, 2009: Simulations of 20th and 21st century Arctic
2 cloud amount in the global climate models assessed in the IPCC AR4. *Climate Dynamics*, **33**, 1099-
3 1115.
- 4 Verlinde, J., and Coauthors, 2007: The mixed-phase Arctic cloud experiment. *Bulletin of the American
5 Meteorological Society*, **88**, 205-221.
- 6 Viana, M., W. Maenhaut, X. Chi, X. Querol, and A. Ištáuey, 2007: Comparative chemical mass closure of
7 fine and coarse aerosols at two sites in South and West Europe: implications for EU air pollution
8 policies. *Atmospheric Environment*, **41**, 315-326.
- 9 Viana, M., X. Chi, W. Maenhaut, X. Querol, A. Ištáuey, P. Mikuska, and Z. Vecera, 2006: Organic and
10 elemental carbon concentrations during summer and winter sampling campaigns in Barcelona, Spain.
11 *Atmospheric Environment*, **40**, 2180-2193.
- 12 Viana, M., and Coauthors, 2008: Characterising exposure to PM aerosols for an epidemiological study.
13 *Atmos. Environ.*, **42**, 1552-1568.
- 14 Vignati, E., M. Karl, M. Krol, J. Wilson, P. Stier, and F. Cavalli, 2010: Sources of uncertainties in modelling
15 black carbon at the global scale. *Atmospheric Chemistry and Physics*, 2595-2611.
- 16 Vlahos, P., and E. Monahan, 2009: A generalized model for the air-sea transfer of dimethyl sulfide at high
17 wind speeds. *Geophysical Research Letters*, ARTN L21605, DOI 10.1029/2009GL040695.
- 18 Vogelmann, A. M., T. P. Ackerman, and R. P. Turco, 1992: Enhancements in biologically effective
19 ultraviolet radiation following volcanic eruptions. *Nature*, **359**, 47-49.
- 20 Volkamer, R., and Coauthors, 2006: Secondary organic aerosol formation from anthropogenic air pollution:
21 Rapid and higher than expected. *Geophysical Research Letters*, ARTN L17811, DOI
22 10.1029/2006GL026899.
- 23 Volodin, E. M., 2008: Relation between temperature sensitivity to doubled carbon dioxide and the
24 distribution of clouds in current climate models. *Izvestiya Atmospheric and Oceanic Physics*, **44**, 288-
25 299.
- 26 von Blohn, N., S. K. Mitra, K. Diehl, and S. Borrmann, 2005: The ice nucleating ability of pollen: Part III:
27 New laboratory studies in immersion and contact freezing modes including more pollen types.
28 *Atmospheric Research*, **78**, 182-189.
- 29 Wagner, R., O. Mohler, H. Saathoff, M. Schnaiter, and T. Leisner, 2010: High variability of the
30 heterogeneous ice nucleation potential of oxalic acid dihydrate and sodium oxalate. *Atmospheric
31 Chemistry and Physics*, **10**, 7617-7641.
- 32 ———, 2011: New cloud chamber experiments on the heterogeneous ice nucleation ability of oxalic acid in
33 the immersion mode. *Atmospheric Chemistry and Physics*, **11**, 2083-2110.
- 34 Wang, B. B., and D. A. Knopf, 2011: Heterogeneous ice nucleation on particles composed of humic-like
35 substances impacted by O(3). *Journal of Geophysical Research-Atmospheres*, **116**.
- 36 Wang, G., H. Wang, Y. Yu, S. Gao, J. Feng, S. Gao, and L. Wang, 2003: Chemical characterization of
37 water-soluble components of PM10 and PM2.5 atmospheric aerosols in five locations of Nanjing,
38 China. *Atmospheric Environment*, **37**, 2893-2902.
- 39 Wang, H., and D. Shooter, 2001: Water soluble ions of atmospheric aerosols in three New Zealand cities:
40 seasonal changes and sources. *Atmospheric Environment*, **35**.
- 41 Wang, H., and G. Feingold, 2009: Modeling mesoscale cellular structures and drizzle in marine
42 stratocumulus. Part II: The microphysics and dynamics of the boundary region between open and
43 closed cells. *Journal of the Atmospheric Sciences*, **66**, 3257-3275.
- 44 Wang, H., K. Kawamura, and D. Shooter, 2005a: Carbonaceous and ionic components in wintertime
45 atmospheric aerosols from two New Zealand cities: Implications for solid fuel combustion.
46 *Atmospheric Environment*, **39**, 5865-5875.
- 47 Wang, H., P. J. Rasch, and G. Feingold, 2011a: Manipulating marine stratocumulus cloud amount and
48 albedo: a process-modelling study of aerosol-cloud-precipitation interactions in response to injection
49 of cloud condensation nuclei. *Atmos. Chem. Phys.*, **11**, 4237-4249.
- 50 Wang, H., G. Feingold, R. Wood, and J. Kazil, 2010a: Modelling microphysical and meteorological controls
51 on precipitation and cloud cellular structures in Southeast Pacific stratocumulus. *Atmospheric
52 Chemistry and Physics*, **10**, 6347-6362.
- 53 Wang, L., A. Khalizov, J. Zheng, W. Xu, Y. Ma, V. Lal, and R. Zhang, 2010b: Atmospheric nanoparticles
54 formed from heterogeneous reactions of organics. *Nature Geoscience*, **3**, 238-242.
- 55 Wang, M., and J. Penner, 2009: Aerosol indirect forcing in a global model with particle nucleation.
56 *Atmospheric Chemistry and Physics*, **9**, 239-260.

- 1 Wang, M., J. Penner, and X. Liu, 2009: Coupled IMPACT aerosol and NCAR CAM3 model: Evaluation of
2 predicted aerosol number and size distribution. *Journal of Geophysical Research-Atmospheres*, ARTN
3 D06302, DOI 10.1029/2008JD010459.
- 4 Wang, M., and Coauthors, 2011b: Aerosol indirect effects in a multi-scale aerosol-climate model PNNL-
5 MMF. *Atmospheric Chemistry and Physics*, **11**, 5431-5455.
- 6 Wang, Y., G. Zhuang, A. Tang, H. Yuan, Y. Sun, S. Chen, and A. Zheng, 2005b: The ion chemistry and the
7 source of PM 2.5 aerosol in Beijing. *Atmospheric Environment*, **39**, 3771-3784.
- 8 Wang, Y., and Coauthors, 2006: The ion chemistry, seasonal cycle, and sources of PM2.5 and TSP aerosol in
9 Shanghai. *Atmospheric Environment*, **40**, 2935-2952.
- 10 Waquet, F., J. Riedi, L. Labonnote, P. Goloub, B. Cairns, J. Deuzé, and D. Tanré, 2009: Aerosol Remote
11 Sensing over Clouds Using A-Train Observations. *Journal of the Atmospheric Sciences*, **66**, 2468-
12 2480.
- 13 Warneke, C., and Coauthors, 2009: Biomass burning in Siberia and Kazakhstan as an important source for
14 haze over the Alaskan Arctic in April 2008. *Geophysical Research Letters*, ARTN L02813, DOI
15 10.1029/2008GL036194.
- 16 Warneke, C., and Coauthors, 2010: An important contribution to springtime Arctic aerosol from biomass
17 burning in Russia. *Geophysical Research Letters*, ARTN L01801, DOI 10.1029/2009GL041816.
- 18 Webb, M., F. H. Lambert, and J. Gregory, 2012: Origins of differences in climate sensitivity, forcing and
19 feedback in climate models., Manuscript submitted to *Climate Dynamics*, 2 November 2011. ed.
- 20 Weinstein, J. P., S. R. Hedges, and S. Kimbrough, 2010: Characterization and aerosol mass balance of
21 PM2.5 and PM10 collected in Conakry, Guinea during the 2004 Harmattan period. *Chemosphere*, **78**,
22 980-988.
- 23 Welti, A., F. Lüönd, O. Stetzer, and U. Lohmann, 2009: Influence of particle size on the ice nucleating
24 ability of mineral dusts. *Atmospheric Chemistry and Physics*, **9**, 6705-6715.
- 25 Wen, G., A. Marshak, R. F. Cahalan, L. A. Remer, and R. G. Kleidman, 2007: 3-D aerosol-cloud radiative
26 interaction observed in collocated MODIS and ASTER images of cumulus cloud fields. *Journal of*
27 *Geophysical Research-Atmospheres*, **112**.
- 28 Wetherald, R. T., and S. Manabe, 1980: Cloud cover and climate sensitivity. *Journal of the Atmospheric*
29 *Sciences*, **37**, 1485-1510.
- 30 Wex, H., G. McFiggans, S. Henning, and F. Stratmann, 2010: Influence of the external mixing state of
31 atmospheric aerosol on derived CCN number concentrations. *Geophysical Research Letters*, **37**, -.
- 32 Wexler, A. S., and S. L. Clegg, 2002: Atmospheric aerosol models for systems including the ions H^+ , NH_4^+ ,
33 Na^+ , SO_4^{2-} , NO_3^- , Cl^- , Br^- , and H_2O . *Journal of Geophysical Research*, **107**, 4207.
- 34 Whitlock, C., D. Bartlett, and E. Gurganus, 1982: Sea foam reflectance and influence on optimum
35 wavelength for remote-sensing of ocean aerosols. *Geophysical Research Letters*, **9**, 719-722.
- 36 Wilcox, E. M., 2010: Stratocumulus cloud thickening beneath layers of absorbing smoke aerosol.
37 *Atmospheric Chemistry and Physics*, **10**, 11769-11777.
- 38 Wild, M., and B. Liepert, 2010: The Earth radiation balance as driver of the global hydrological cycle.
39 *Environ. Res. Lett.*, **5**, 025203.
- 40 Williams, K. D., and G. Tselioudis, 2007: GCM intercomparison of global cloud regimes: present-day
41 evaluation and climate change response. *Climate Dynamics*, **29**, 231-250.
- 42 Williams, K. D., and M. J. Webb, 2009: A quantitative performance assessment of cloud regimes in climate
43 models. *Climate Dynamics*, **33**, 141-157.
- 44 Williams, K. D., A. Jones, D. L. Roberts, C. A. Senior, and M. J. Woodage, 2001: The response of the
45 climate system to the indirect effects of anthropogenic sulfate aerosol. *Climate Dynamics*, **17**, 845-
46 856.
- 47 Williams, K. D., and Coauthors, 2006: Evaluation of a component of the cloud response to climate change in
48 an intercomparison of climate models. *Climate Dynamics*, **26**, 145-165.
- 49 Winker, D. M., and Coauthors, 2009: Overview of the CALIPSO mission and CALIOP data processing
50 algorithms. *Journal of Atmospheric and Oceanic Technology*, **26**, 2310-2323.
- 51 Wise, M. E., K. J. Baustian, and M. A. Tolbert, 2009: Laboratory studies of ice formation pathways from
52 ammonium sulfate particles. *Atmospheric Chemistry and Physics*, **9**, 1639-1646.
- 53 Wise, M. E., K. J. Baustian, and M. A. Tolbert, 2010: Internally mixed sulfate and organic particles as
54 potential ice nuclei in the tropical tropopause region. *Proceedings of the National Academy of*
55 *Sciences of the United States of America*, **107**, 6693-6698.

- 1 Wolters, E. L. A., H. M. Deneke, B. van den Hurk, J. F. Meirink, and R. A. Roebeling, 2010: Broken and
2 inhomogeneous cloud impact on satellite cloud particle effective radius and cloud-phase retrievals.
3 *Journal of Geophysical Research-Atmospheres*, **115**, 14.
- 4 Wong, S., and A. E. Dessler, 2005: Suppression of deep convection over the tropical North Atlantic by the
5 Saharan Air Layer. *Geophysical Research Letters*, **32**, L09808.
- 6 Wong, S., A. E. Dessler, N. M. Mahowald, P. Yang, and Q. Feng, 2009: Maintenance of lower tropospheric
7 temperature inversion in the Saharan Air Layer by dust and dry anomaly. *Journal of Climate*, **22**,
8 5149-5162.
- 9 Wood, R., 2005: Drizzle in stratiform boundary layer clouds. Part II: Microphysical aspects. *Journal of the*
10 *Atmospheric Sciences*, **62**, 3034-3050.
- 11 Wood, R., 2007: Cancellation of aerosol indirect effects in marine stratocumulus through cloud thinning.
12 *Journal of the Atmospheric Sciences*, **64**, 2657-2669.
- 13 Wood, R., and C. S. Bretherton, 2006: On the relationship between stratiform low cloud cover and lower-
14 tropospheric stability. *Journal of Climate*, **19**, 6425-6432.
- 15 Wood, R., and Coauthors, 2011: The VAMOS Ocean-Cloud-Atmosphere-Land Study Regional Experiment
16 (VOCALS-REx): goals, platforms, and field operations. *Atmospheric Chemistry and Physics*, **11**, 627-
17 654.
- 18 Woodhouse, M., G. Mann, K. Carslaw, and O. Boucher, 2008: New Directions: The impact of oceanic iron
19 fertilisation on cloud condensation nuclei. *Atmospheric Environment*, 5728-5730.
- 20 Woodhouse, M., K. Carslaw, G. Mann, S. Vallina, M. Vogt, P. Halloran, and O. Boucher, 2010: Low
21 sensitivity of cloud condensation nuclei to changes in the sea-air flux of dimethyl-sulphide.
22 *Atmospheric Chemistry and Physics*, 7545-7559.
- 23 Woodward, S., D. Roberts, and R. Betts, 2005: A simulation of the effect of climate change-induced
24 desertification on mineral dust aerosol. *Geophysical Research Letters*, ARTN L18810, DOI
25 10.1029/2005GL023482.
- 26 Wyant, M. C., C. S. Bretherton, and P. N. Blossey, 2009: Subtropical low cloud response to a warmer
27 climate in a superparameterized climate model: Part I, Regime sorting and physical mechanisms. *J.*
28 *Adv. Modeling Earth Syst.*, **1**, 7.
- 29 Wyant, M. C., C. S. Bretherton, P. N. Blossey, and M. Khairoutdinov, 2011: Fast cloud adjustment to
30 increasing CO₂ in a superparameterized climate model. *J. Adv. Model. Earth Systems*, **in press**.
- 31 Wyant, M. C., and Coauthors, 2006: A comparison of low-latitude cloud properties and their response to
32 climate change in three AGCMs sorted into regimes using mid-tropospheric vertical velocity. *Climate*
33 *Dynamics*, **27**, 261-279.
- 34 Xiao, H.-Y., and C.-Q. Liu, 2004: Chemical characteristics of water-soluble components in TSP over
35 Guiyang, SW China, 2003. *Atmospheric Environment*, **38**, 6297-6306.
- 36 Xu, K. M., A. N. Cheng, and M. H. Zhang, 2010: Cloud-resolving simulation of low-cloud feedback to an
37 increase in sea surface temperature. *Journal of the Atmospheric Sciences*, **67**, 730-748.
- 38 Xue, H., G. Feingold, and B. Stevens, 2008: Aerosol effects on clouds, precipitation, and the organization of
39 shallow cumulus convection. *Journal of the Atmospheric Sciences*, **65**, 392-406.
- 40 Yankofsky, S. A., Z. Levin, T. Bertold, and N. Sandlerman, 1981: Some basic characteristics of bacterial
41 freezing nuclei. *Journal of Applied Meteorology*, **20**, 1013-1019.
- 42 Yao, X., and Coauthors, 2002: The water-soluble ionic composition of PM_{2.5} in Shanghai and Beijing,
43 China. *Atmospheric Environment*, **36**, 4223-4234.
- 44 Ye, B., and Coauthors, 2003: Concentration and chemical composition of PM_{2.5} in Shanghai for a 1-year
45 period. *Atmospheric Environment*, **37**, 499-510.
- 46 Yin, J., and R. M. Harrison, 2008: Pragmatic mass closure study for PM_{1.0}, PM_{2.5} and PM₁₀ at roadside,
47 urban background and rural sites. *Atmos. Environ.*, **42**, 980-988.
- 48 Yin, J. H., 2005: A consistent poleward shift of the storm tracks in simulations of 21st century climate.
49 *Geophysical Research Letters*, **32**, 4.
- 50 Yokohata, T., M. J. Webb, M. Collins, K. D. Williams, M. Yoshimori, J. C. Hargreaves, and J. D. Annan,
51 2010: Structural similarities and differences in climate responses to CO₂ increase between two
52 perturbed physics ensembles. *Journal of Climate*, **23**, 1392-1410.
- 53 Yokohata, T., and Coauthors, 2008: Comparison of equilibrium and transient responses to CO₂ increase in
54 eight state-of-the-art climate models. *Tellus*, **60A**, 946-961.
- 55 Yoshimori, M., and A. J. Broccoli, 2008: Equilibrium response of an atmosphere-mixed layer ocean model
56 to different radiative forcing agents: Global and zonal mean response. *Journal of Climate*, **21**, 4399-
57 4423.

- 1 Yoshimori, M., T. Yokohata, and A. Abe-Ouchi, 2009: A comparison of climate feedback strength between
2 CO₂ doubling and LGM experiments. *Journal of Climate*, **22**, 3374-3395.
- 3 Young, I. R., S. Zieger, and A. V. Babanin, 2011: Global trends in wind speed and wave height. *Science*,
4 **332**, 451-455.
- 5 Yttri, K. E., 2007: Concentrations of particulate matter (PM₁₀, PM_{2.5}) in Norway. Annual and seasonal
6 trends and spatial variability Annex A, EMEP Particulate Matter Assessment Report, Part B, report
7 EMEP/CCC-Report 8/2007, 292-307, ref. O-7726.
- 8 Yu, F., 2010: Ion-mediated nucleation in the atmosphere: Key controlling parameters, implications, and
9 look-up table. *Journal of Geophysical Research-Atmospheres*, ARTN D03206, DOI
10 10.1029/2009JD012630.
- 11 Yu, F., and G. Luo, 2009: Simulation of particle size distribution with a global aerosol model: contribution
12 of nucleation to aerosol and CCN number concentrations. *Atmospheric Chemistry and Physics*, 7691-
13 7710.
- 14 Yu, F., and Coauthors, 2010a: Spatial distributions of particle number concentrations in the global
15 troposphere: Simulations, observations, and implications for nucleation mechanisms. *Journal of*
16 *Geophysical Research-Atmospheres*, ARTN D17205, DOI 10.1029/2009JD013473.
- 17 Yu, H., M. Chin, D. Winker, A. Omar, Z. Liu, C. Kittaka, and T. Diehl, 2010b: Global view of aerosol
18 vertical distributions from CALIPSO lidar measurements and GOCART simulations: Regional and
19 seasonal variations. *Journal of Geophysical Research-Atmospheres*, ARTN D00H30, DOI
20 10.1029/2009JD013364-.
- 21 Yu, H., and Coauthors, 2006: A review of measurement-based assessments of the aerosol direct radiative
22 effect and forcing. *Atmospheric Chemistry and Physics*, 613-666.
- 23 Yuan, T., L. A. Remer, and H. Yu, 2011: Microphysical, macrophysical and radiative signatures of volcanic
24 aerosols in trade wind cumulus observed by the A-Train. *Atmos. Chem. Phys.*, **11**, 7119-7132.
- 25 Zarzycki, C. M., and T. C. Bond, 2010: How much can the vertical distribution of black carbon affect its
26 global direct radiative forcing? *Geophys. Res. Lett.*, **37**, L20807.
- 27 Zaveri, R. A., R. C. Easter, J. D. Fast, and L. K. Peters, 2008: Model for Simulating Aerosol Interactions and
28 Chemistry (MOSAIC). *Journal of Geophysical Research*, **113**, D13204.
- 29 Zelinka, M. D., and D. L. Hartmann, 2010: Why is longwave cloud feedback positive? *Journal of*
30 *Geophysical Research*, **115**, D16117.
- 31 Zelinka, M. D., S. A. Klein, and D. L. Hartmann, 2011a: Computing and partitioning cloud feedbacks using
32 cloud property histograms. Part I: Cloud radiative kernels. *J. Climate*, **in press**.
- 33 —, 2011b: Computing and partitioning cloud feedbacks using cloud property histograms. Part II:
34 Attribution to changes in cloud amount, altitude, and optical depth. *Journal of Climate*, **in press**.
- 35 Zhang, G. J., A. M. Vogelmann, M. P. Jensen, W. D. Collins, and E. P. Luke, 2010a: Relating satellite-
36 observed cloud properties from MODIS to meteorological conditions for marine boundary layer
37 clouds. *Journal of Climate*, **23**, 1374-1391.
- 38 Zhang, K., H. Wan, B. Wang, M. Zhang, J. Feichter, and X. Liu, 2010b: Tropospheric aerosol size
39 distributions simulated by three online global aerosol models using the M7 microphysics module.
40 *Atmospheric Chemistry and Physics*, **10**, 6409-6434.
- 41 Zhang, M. H., and C. Bretherton, 2008: Mechanisms of low cloud-climate feedback in idealized single-
42 column simulations with the Community Atmospheric Model, version 3 (CAM3). *Journal of Climate*,
43 **21**, 4859-4878.
- 44 Zhang, Q., D. R. Worsnop, M. R. Canagaratna, and J. L. Jimenez, 2005a: Hydrocarbon-like and oxygenated
45 organic aerosols in Pittsburgh: insights into sources and processes of organic aerosols. *Atmospheric*
46 *Chemistry and Physics*, **5**, 3289-3311.
- 47 Zhang, Q., M. R. Alfarra, D. R. Worsnop, J. D. Allan, H. Coe, M. R. Canagaratna, and J. L. Jimenez, 2005b:
48 Deconvolution and quantification of hydrocarbon-like and oxygenated organic aerosols based on
49 aerosol mass spectrometry. *Environmental Science and Technology*, **39**, 4938-4952.
- 50 Zhang, Q., and Coauthors, 2007: Ubiquity and dominance of oxygenated species in organic aerosols in
51 anthropogenically-influenced Northern Hemisphere midlatitudes. *Geophysical Research Letters*,
52 ARTN L13801, DOI 10.1029/2007GL029979.
- 53 Zhang, X. Y., R. Arimoto, Z. S. An, J. J. Cao, and D. Wang, 2001: Atmospheric dust aerosol over the
54 Tibetan Plateau. *Journal of Geophysical Research*, **106**, 18471-18476.
- 55 Zhang, X. Y., Y. Q. Wang, X. C. Zhang, W. Guo, and S. L. Gong, 2008: Carbonaceous aerosol composition
56 over various regions of China during 2006. *J. Geophys. Res.*, **113**, D14111.

- 1 Zhang, X. Y., J. J. Cao, L. M. Li, R. Arimoto, Y. Cheng, B. Huebert, and D. Wang, 2002: Characterization
2 of atmospheric aerosol over Xian in the south margin of the loess plateau, China. *Atmospheric*
3 *Environment*, **36**, 4189-4199.
- 4 Zhang, X. Y., Y. Q. Wang, T. Niu, X. C. Zhang, S. L. Gong, Y. M. Zhang, and J. Y. Sun, 2011a:
5 Atmospheric aerosols in China: spatial/temporal variability, chemical signature, regional haze
6 distribution and comparisons with global aerosols. *Atmos. Chem. Phys. Discuss.*, **11**, 26571-26615.
- 7 Zhang, Y., and Coauthors, 2004: Development and application of the Model of Aerosol Dynamics, Reaction,
8 ionization, and Dissolution (MADRID). *Journal of Geophysical Research*, **109**, D01202.
- 9 Zhang, Y. M., X. Y. Zhang, J. Y. Sun, W. L. Lin, S. L. Gong, X. J. Shen, and S. Yang, 2011b:
10 Characterization of new particle and secondary aerosol formation during summertime in Beijing,
11 China. *Tellus*, **63B**, 382-394.
- 12 Zhao, T. L., S. L. Gong, X. Y. Zhang, A. A. Mawgoud, and Y. P. Shao, 2006: An assessment of dust
13 emission schemes in modeling east Asian dust storms. *Journal of Geophysical Research*, **111**,
14 D05S90.
- 15 Zhao, T. X. P., H. B. Yu, I. Laszlo, M. Chin, and W. C. Conant, 2008: Derivation of component aerosol
16 direct radiative forcing at the top of atmosphere for clear-sky oceans. *Journal of Quantitative*
17 *Spectroscopy & Radiative Transfer*, **109**, 1162-1186.
- 18 Zhuang, B., L. Liu, F. Shen, T. Wang, and Y. Han, 2010: Semidirect radiative forcing of internal mixed
19 black carbon cloud droplet and its regional climatic effect over China. *Journal of Geophysical*
20 *Research-Atmospheres*, **115**, -.
- 21 Zimmermann, F., S. Weinbruch, L. Schutz, H. Hofmann, M. Ebert, K. Kandler, and A. Worringer, 2008: Ice
22 nucleation properties of the most abundant mineral dust phases. *Journal of Geophysical Research-*
23 *Atmospheres*, **113**.
- 24 Zobrist, B., T. Koop, B. P. Luo, C. Marcolli, and T. Peter, 2007: Heterogeneous ice nucleation rate
25 coefficient of water droplets coated by a nonadecanol monolayer. *Journal of Physical Chemistry C*,
26 **111**, 2149-2155.
- 27 Zobrist, B., and Coauthors, 2006: Oxalic acid as a heterogeneous ice nucleus in the upper troposphere and its
28 indirect aerosol effect. *Atmospheric Chemistry and Physics*, **6**, 3115-3129.
- 29 Zuberi, B., A. K. Bertram, C. A. Cassa, L. T. Molina, and M. J. Molina, 2002: Heterogeneous nucleation of
30 ice in $(\text{NH}_4)_2\text{SO}_4\text{-H}_2\text{O}$ particles with mineral dust immersions. *Geophysical Research Letters*, **29**.
- 31 Zubler, E. M., U. Lohmann, D. Lüthi, C. Schär, and A. Muehlbauer, 2011: Statistical analysis of aerosol
32 effects on simulated mixed-phase clouds and precipitation in the Alps. *J. Atmos. Sci.*, **68**, 1474-1492.
- 33 Zuidema, P., H. Xue, and G. Feingold, 2008: Shortwave radiative impacts from aerosol effects on marine
34 shallow cumuli. *Journal of the Atmospheric Sciences*, **65**, 1979-1990.
- 35
36

1 **Tables**
 2
 3

4 **Table 7.1:** Global and regional anthropogenic emissions important for aerosol formation and tropospheric chemistry. The maximum and minimum values from available inventories
 5 are presented. Units for NO_x are Tg NO yr⁻¹, other units are Tg yr⁻¹. Adapted from Granier et al. (2011).

Year 2000 emissions Tg/year	CO		NO _x		CH ₄		NMVOCs		BC		OC		SO ₂		NH ₃	
	<i>MIN</i>	<i>MAX</i>	<i>MIN</i>	<i>MAX</i>	<i>MIN</i>	<i>MAX</i>	<i>MIN</i>	<i>MAX</i>	<i>MIN</i>	<i>MAX</i>	<i>MIN</i>	<i>MAX</i>	<i>MIN</i>	<i>MAX</i>	<i>MIN</i>	<i>MAX</i>
Total	467.50	610.80	58.70	68.60	275.20	310.30	121.00	139.50	4.60	5.60	6.40	12.70	102.00	145.00	37.50	38.90
Western Europe	21.40	35.40	5.90	9.00	16.30	22.00	9.20	14.30	0.32	0.38	0.32	0.40	6.10	14.10	3.40	4.50
Central Europe	7.80	12.30	1.60	1.90	6.10	7.70	2.30	3.50	0.11	0.21	0.25	0.39	4.60	10.00	1.10	1.20
USA	55.90	94.40	11.50	14.10	26.20	40.70	13.00	17.50	0.27	0.40	0.36	0.51	13.50	17.80	3.30	4.40
Canada	4.20	11.20	1.20	1.70	3.90	5.00	1.50	3.40	0.04	0.04	0.03	0.06	2.20	2.90	0.51	0.60
Central America	10.00	15.10	1.50	2.10	8.40	9.10	2.90	4.10	0.11	0.11	0.17	0.35	3.70	4.10	1.10	1.10
South America	22.30	26.50	2.80	3.80	26.40	30.00	8.40	12.90	0.20	0.33	0.32	0.83	3.80	8.80	3.40	3.50
Africa	49.40	83.20	2.70	5.90	25.00	29.40	10.80	14.50	0.46	0.62	1.05	1.91	5.30	8.80	2.30	2.40
China	95.50	137.30	6.90	9.80	33.10	49.40	11.50	24.50	0.71	1.41	1.10	3.80	19.20	21.10	8.90	13.60
India	40.30	79.40	2.70	4.90	25.70	33.80	7.30	10.80	0.45	0.84	1.00	3.27	4.00	7.90	3.70	8.50
Oceania	2.60	5.70	1.10	1.90	6.40	6.80	0.00	1.50	0.03	0.04	0.04	0.08	2.40	2.70	0.72	0.72

1 **Table 7.2:** Key aerosol properties of the main aerosol species in the troposphere. Brown carbon is a particular type of OA but is treated here as an additional component because it is
 2 light absorbing. The estimate of aerosol burdens and lifetimes in the troposphere are based on the AeroCom models.

Aerosol Species	Global Burden	Mass Size Distribution	Sources	Sinks	Lifetime	Key Climate Relevant Properties
Black carbon		Freshly emitted: 0-80 nm Aged: accumulation mode	Combustion of fossil fuels, biofuels and biomass	Wet deposition Dry deposition	7–10 days	Large mass absorption efficiency in the visible
Brown carbon		Freshly emitted: 100-400 nm Aged: accumulation mode	Combustion of biofuels and biomass	Wet deposition Dry deposition	1 week	Medium mass absorption efficiency in the visible. Light scattering.
Organic aerosol		POA: Aitken mode SOA: nuclei mode Aged OA : accumulation mode Biogenic POA : coarse mode	Combustion of fossil fuel, biofuel and biomass. Continental and marine ecosystems. Some anthropogenic non-combustion activities.	Wet deposition Dry deposition	1 week	Light scattering. Lens effect when deposited on black or brown carbon. CCN active (depending on aging time and mechanism). IN active (biogenic POA)
Sulphate		Secondary: Nuclei, Aitken, and accumulation mode Primary: coarse mode	Primary: marine and volcanic emissions. Secondary: oxidation of SO ₂ from natural and anthropogenic sources	Wet deposition Dry deposition	1 week	Light scattering. Very hygroscopic. Lens effect when deposited on black or brown carbon. CCN active.
Nitrate		Accumulation and coarse modes	Oxidation of NO _x	Wet deposition Dry deposition	1 week	Light scattering. CCN active.
Dust	(sensitive to size cutoff)	Coarse and super-coarse modes, with a small accumulation mode	Wind erosion, soil resuspension. Some agricultural practices and industrial activities (cement)	Sedimentation Dry deposition Wet deposition	1 day to 1 week depending on size	IN active, light scattering and absorption, greenhouse effect.
Sea-salt	(sensitive to size cutoff)	Coarse mode and accumulation mode	Wave breaking. Wind erosion.	Sedimentation Wet deposition Dry deposition	1 day to 1 week depending on size	Light scattering. Very hygroscopic. CCN active.

3

4

Chapter 7: Clouds and Aerosols

Coordinating Lead Authors: Olivier Boucher (France), David Randall (USA)

Lead Authors: Paulo Artaxo (Brazil), Christopher Bretherton (USA), Graham Feingold (USA), Piers Forster (UK), Veli-Matti Kerminen (Finland), Yutaka Kondo (Japan), Hong Liao (China), Ulrike Lohmann (Switzerland), Philip Rasch (USA), S. K. Satheesh (India), Steven Sherwood (Australia), Bjorn Stevens (Germany), Xiao-Ye Zhang (China)

Contributing Authors: Govindswamy Bala (India), Nicolas Bellouin (UK), Cristina Facchini (Italy), Mark Flanner (USA), Steve Ghan (USA), Claire Granier (France), Corinna Hoose (Germany), Makoto Koike (Japan), Natalie Mahowald (USA), Gunnar Myhre (Norway), Alan Robock (USA), Bjørn Samset (Norway), Hauke Schmidt (Germany), Michael Schulz (Norway), Trude Storelvmo (USA)

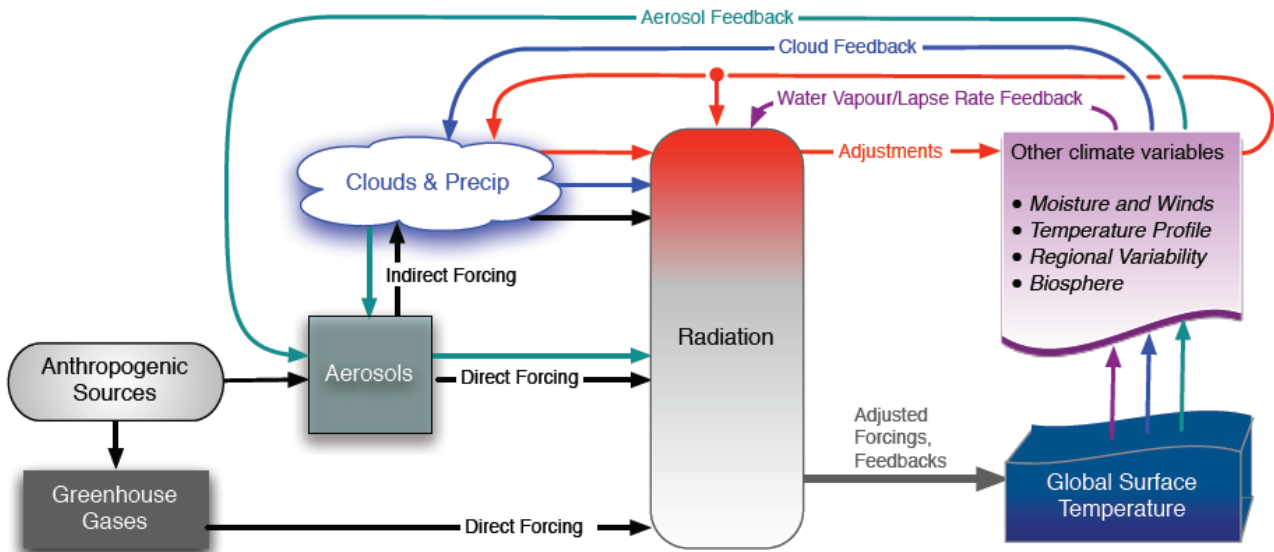
Review Editors: Sandro Fuzzi (Italy), Joyce Penner (USA), Venkatachalam Ramaswamy (USA), Claudia Stubenrauch (France)

Date of Draft: 16 December 2011

Notes: TSU Compiled Version

1 **Figures**

2



3

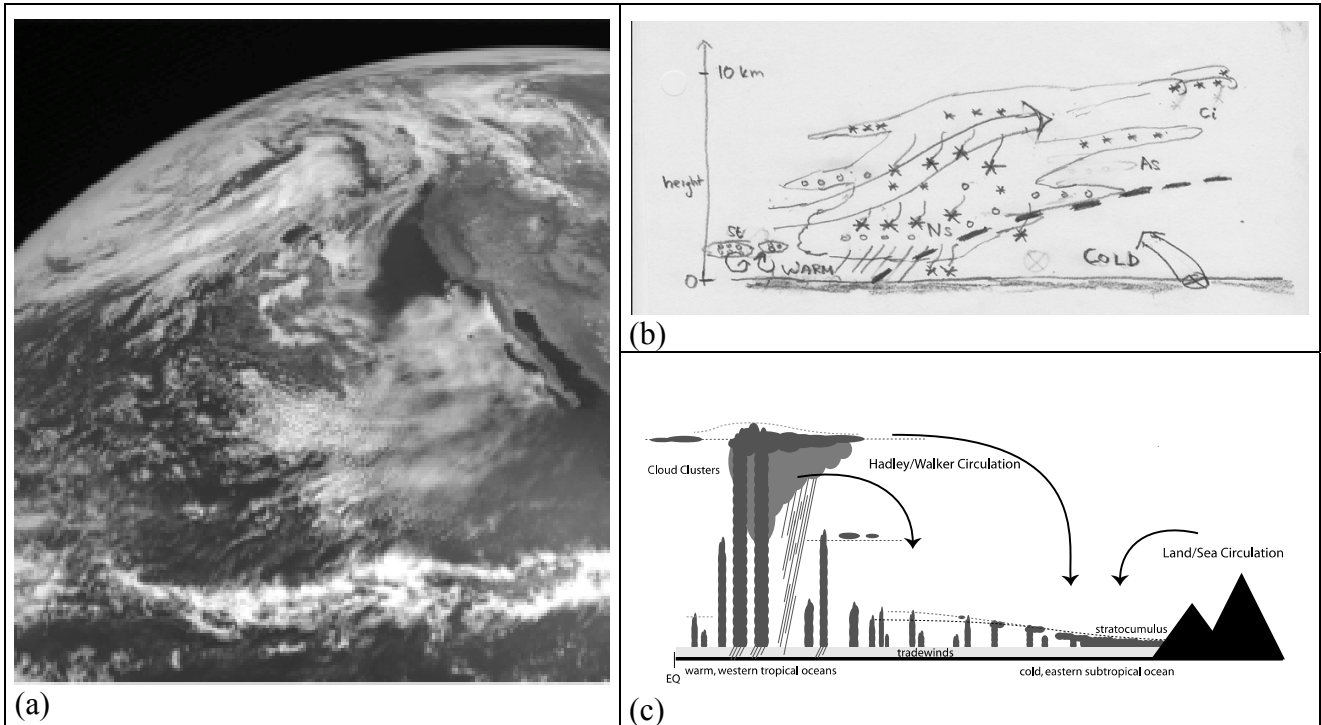
4

5 **Figure 7.1:** Overview of feedback and forcing pathways involving clouds and aerosols. Forcings are represented by
 6 dark arrows; forcing agents are boxes with grey shadows, rapid forcing adjustments (or rapid response) are red arrows
 7 and feedbacks are other-colored arrows. See text for further discussion.

8

9

1



2

3

4

5

6

7

8

9

10

11

12

13

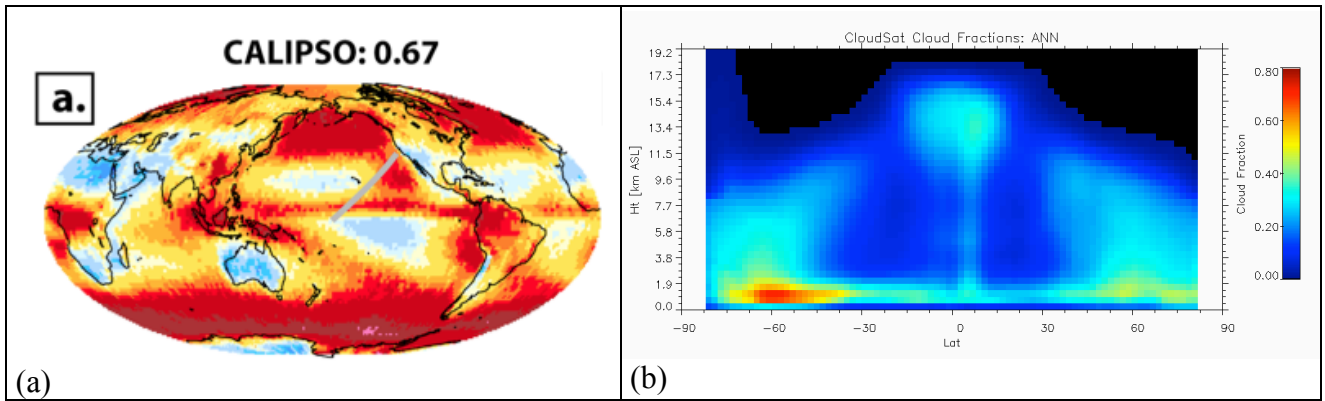
14

15

16

Figure 7.2: Diverse cloud regimes reflect diverse meteorology. (a) A visible-wavelength geostationary satellite image shows (from top to bottom) expanses and long arcs of cloud associated with extratropical cyclones, subtropical coastal stratocumulus near Baja California breaking up into shallow cumulus clouds in the central Pacific, and mesoscale convective systems outlining the Pacific ITCZ. (b) A schematic vertical section through a typical warm front of an extratropical cyclone shows multiple layers of upper-tropospheric ice (cirrus) and mid-tropospheric water (altostratus) cloud upwind of the frontal zone, an extensive region of nimbostratus associated with frontal uplift and turbulence-driven boundary layer cloud in the warm sector. (c) A schematic cross section along the low-level trade wind flow from a subtropical west coast of a continent to the ITCZ shows typical low-latitude cloud types, shallow stratocumulus in the cool waters of the oceanic upwelling zone near the coast, trapped under a strong subsidence inversion, shallow cumulus of warmer waters further offshore and a transition into precipitating cumulonimbus cloud systems with extensive cirrus anvils associated with rising air motions in the ITCZ.

1



2

3

4

5

6

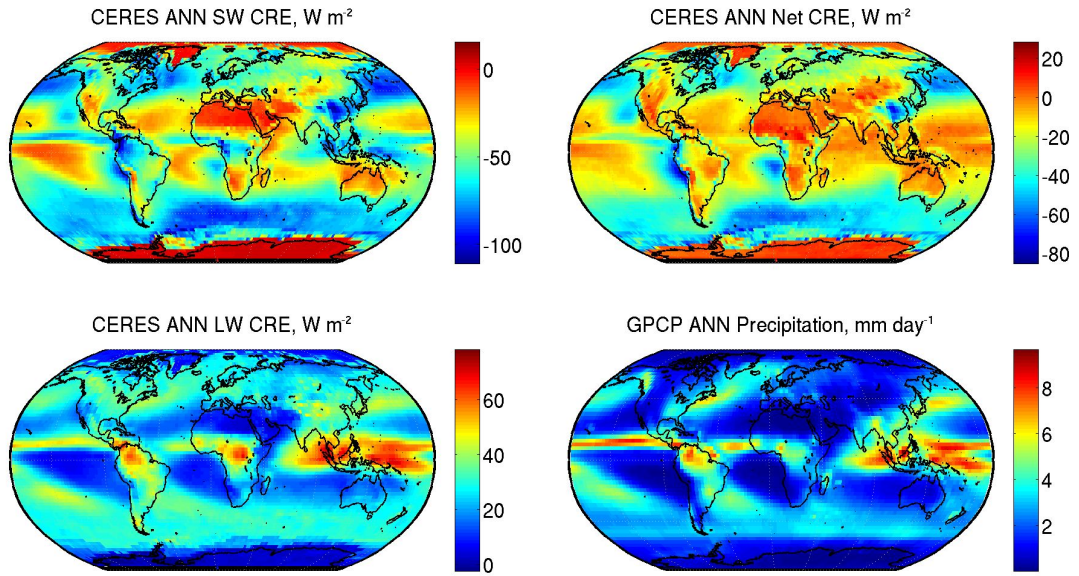
7

8

9

Figure 7.3: Annual-mean cloud fractional occurrence based on four years of satellite observations (June 2006-February 2011) from CloudSat and Calipso (Kay et al., 2011; COSP simulator). (a) Geographical mean, with thin cloud (SR < 5) removed; (b) latitude-height section of zonal mean cloud cover. [PLACEHOLDER FOR SECOND ORDER DRAFT: further graphical refinement.]

1



2

3

4

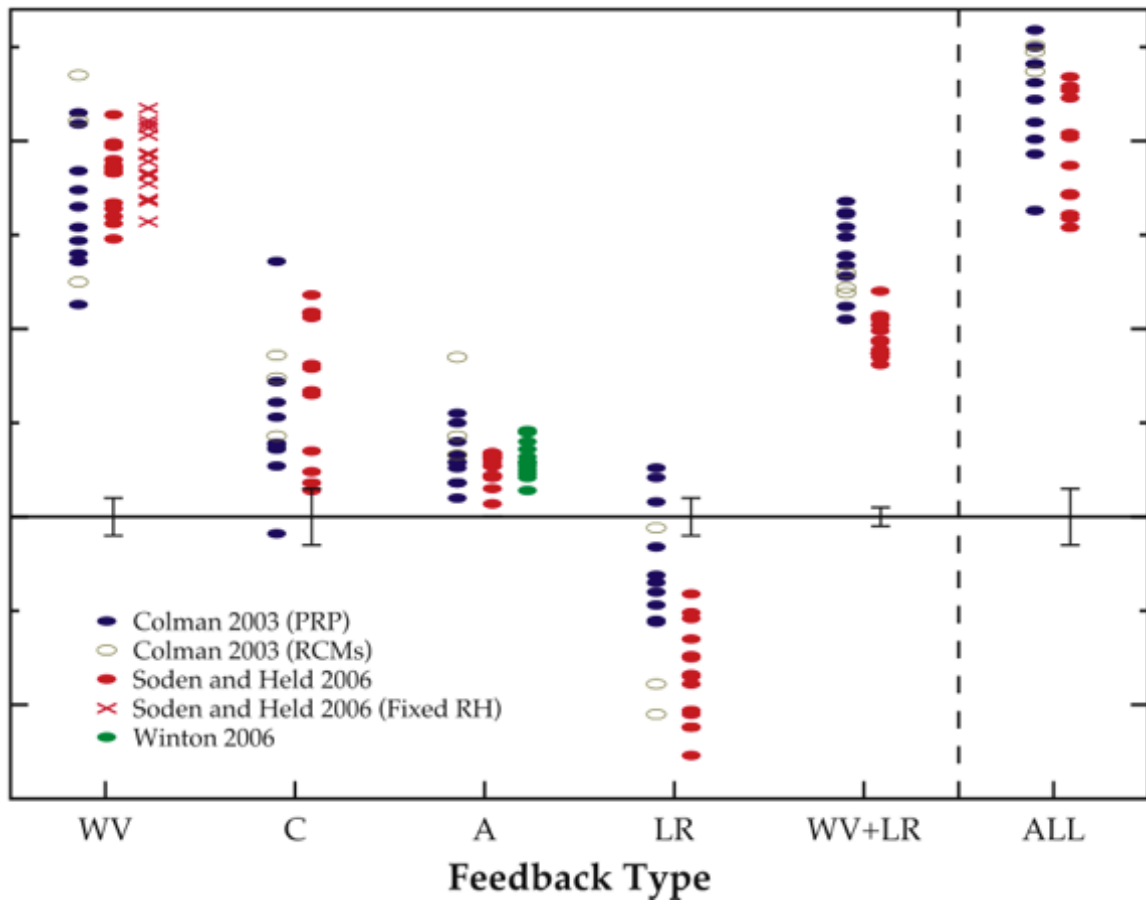
Figure 7.4: Distribution of annual-mean SWCRE, LWCRE, net CRE (from CERES-EBAF) and precipitation (from CMAP).

5

6

7

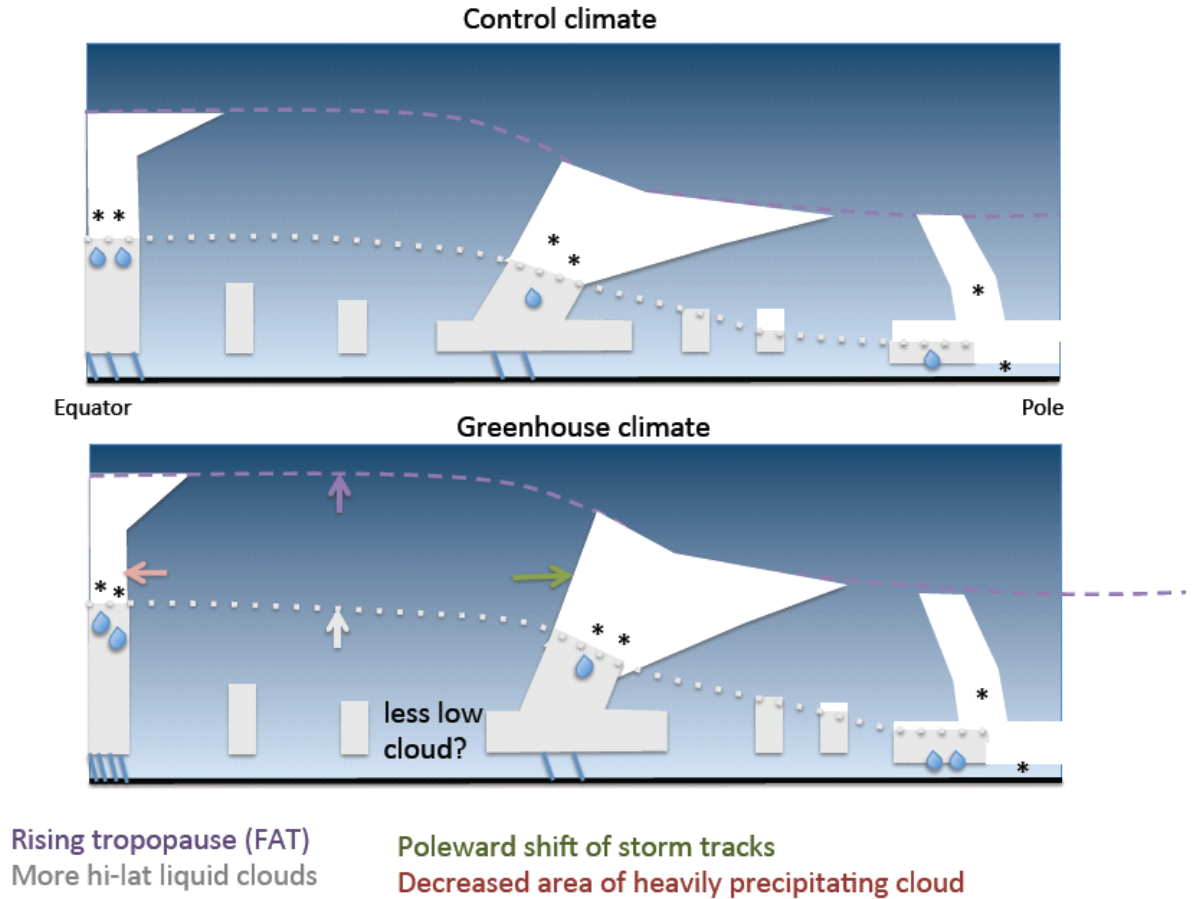
1



2
3
4
5
6
7

Figure 7.5: CFMIP figure on cloud feedbacks in CMIP5 models. [PLACEHOLDER FOR SECOND ORDER DRAFT: CMIP3 version used as placeholder.]

1



2

3

4

5

6

7

8

9

10

11

12

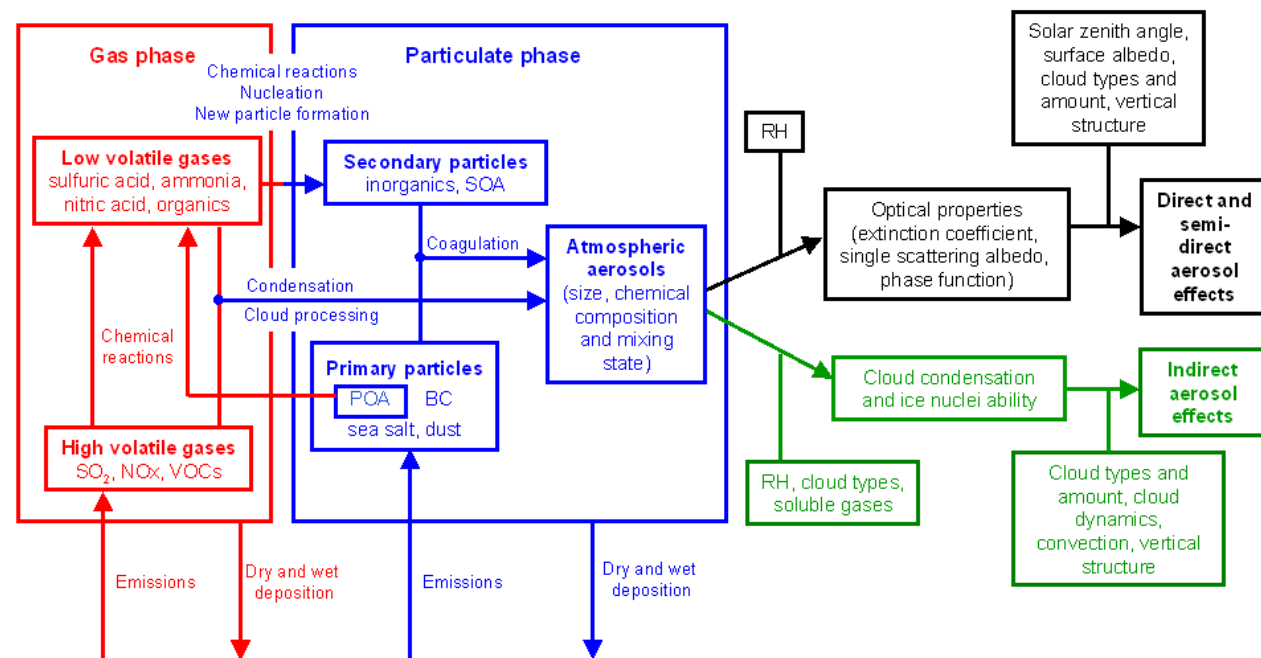
13

14

15

Figure 7.6: Robust cloud responses to greenhouse warming simulated by the CMIP3 multimodel ensemble. Panel (a) is a schematic latitude-altitude section showing typical cloud types in a pre-industrial climate. Grey (white) indicates clouds composed predominantly of liquid water (ice). Raindrops and snowflakes indicate the typical precipitation type. Dotted line indicates the typical freezing level, and purple dashed line indicates the tropopause. Panel (b) shows the same cross section for a warmer climate, with arrows denoting the movement of different boundaries. Tropical deep convection regions narrow and intensify, the subsidence regions of the subtropics widen poleward, with most GCMs projecting low cloud decreases in this area, and storm track cloud and precipitation also shift poleward. Cirrus cloud tops rise in lockstep with the tropopause, helping induce positive longwave cloud feedbacks. The rising freezing level causes more cloud to become liquid, contributing to increased optical thickness of high latitude clouds in the CMIP3 multimodel mean. [PLACHOLDER FOR SECOND ORDER DRAFT: CMIP5]

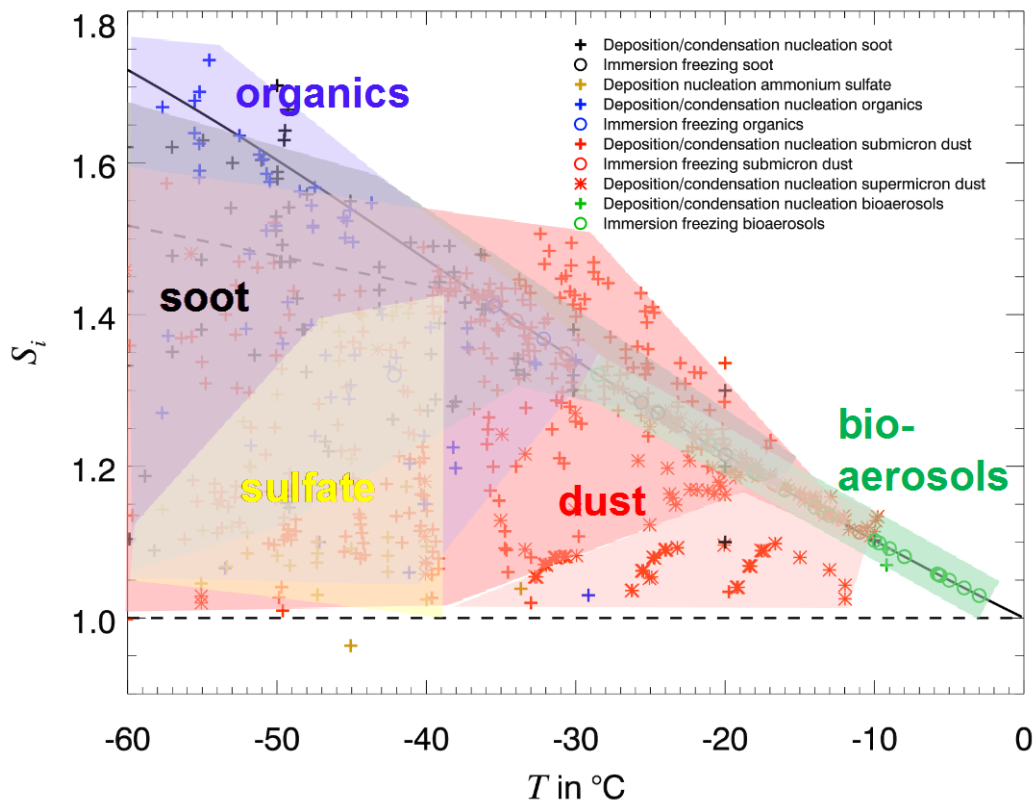
1



2
3
4
5
6
7
8
9

Figure 7.7: Overview of atmospheric aerosol processes and meteorological variables influencing the aerosol semi-direct, direct and indirect aerosol effects. Red designates gas phase processes and variables; blue designates particulate (aerosol) phase processes and variables; processes and variables relevant to the aerosol direct and semi-direct effects appear in black, while those relevant to the aerosol indirect effects appear in green.

1



2

3

4

5

6

7

8

9

10

11

12

13

14

15

16

17

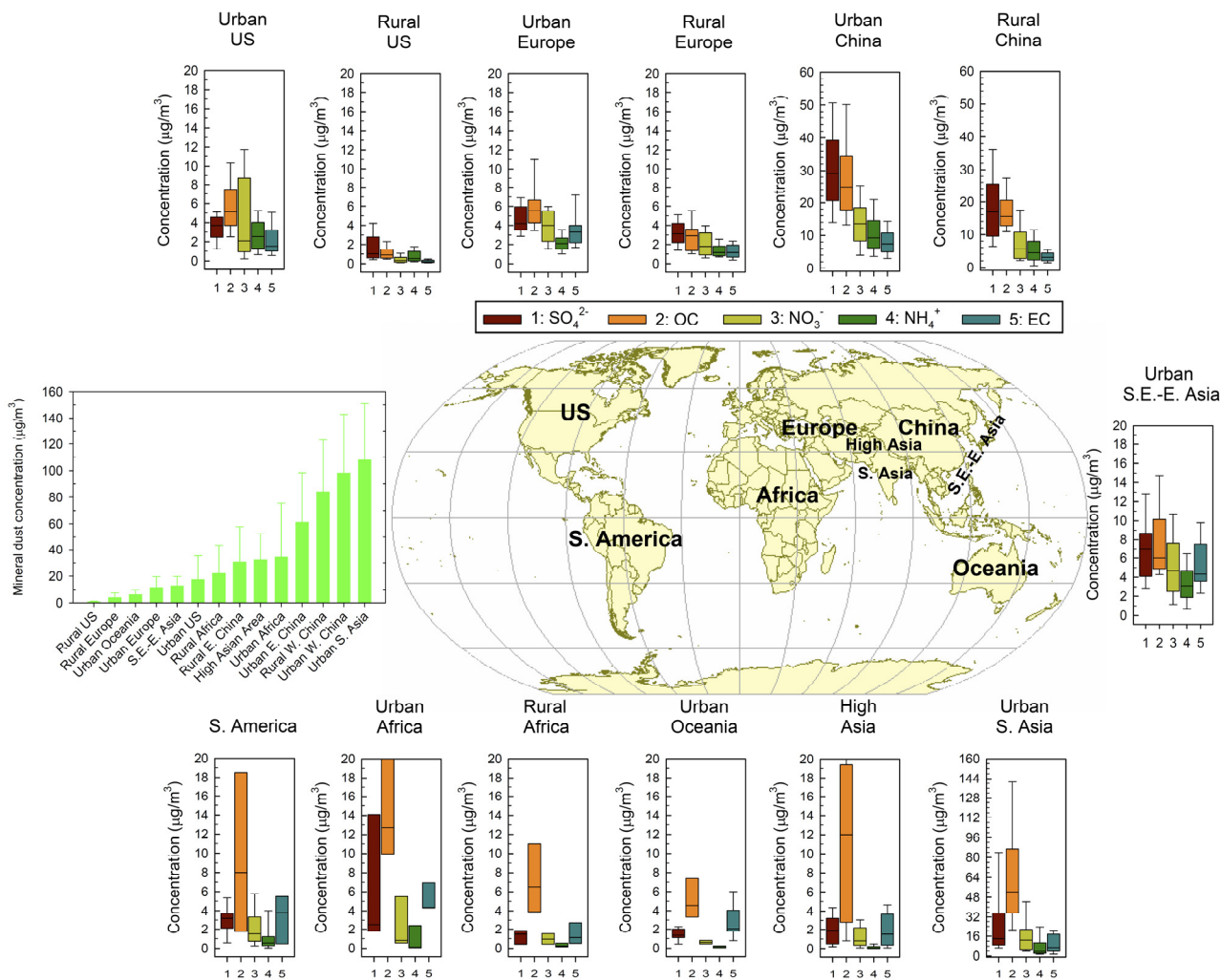
18

19

20

Figure 7.8: The onset temperatures and relative humidities for deposition/condensation freezing and immersion freezing for bioaerosols (Ahern et al., 2007; Diehl et al., 2001; Iannone et al., 2011; Kanji et al., 2011; Mohler et al., 2008; Mortazavi et al., 2008; von Blohn et al., 2005; Yankofsky et al., 1981), mineral dusts (Archuleta et al., 2005; Bundke et al., 2008; Connolly et al., 2009; Cziczo et al., 2009a; Field et al., 2006; Kanji and Abbatt 2006; Kanji et al., 2011; Knopf and Koop 2006; Koehler et al., 2010; Kulkarni and Dobbie 2010; Lüönd et al., 2010; Mohler et al., 2006; Murray et al., 2011; Niedermeier et al., 2010; Niemand et al., 2011; Roberts and Hallett 1968; Salam et al., 2006; Schaller and Fukuta 1979; Welte et al., 2009; Zimmermann et al., 2008), organics (Baustian et al., 2010; Kanji et al., 2008; Petters et al., 2009; Prenni et al., 2007; Shilling et al., 2006; Wagner et al., 2010, 2011; Wang and Knopf 2011; Zobrist et al., 2007), solid ammonium sulphate (Abbatt et al., 2006; Baustian et al., 2010; Mangold et al., 2005; Shilling et al., 2006; Wise et al., 2009; 2010) and BC (soot) (Crawford et al., 2011; DeMott 1990; DeMott et al., 1999; Diehl and Mitra 1998; Dymarska et al., 2006; Fornea et al., 2009; Gorbunov et al., 2001; Kanji et al., 2011; Mohler et al., 2005), from a compilation of experimental data of sub- and super-micron aerosol particles in the literature (for references see supplementary material). The large range of observed ice nucleation onset conditions is due to different experimental setups, particle sizes, activated fractions and chemical composition. Only those IN species for which at least three papers exists are shown. The dashed line refers to the homogeneous freezing of solution droplets after (Koop et al., 2000).

1



2

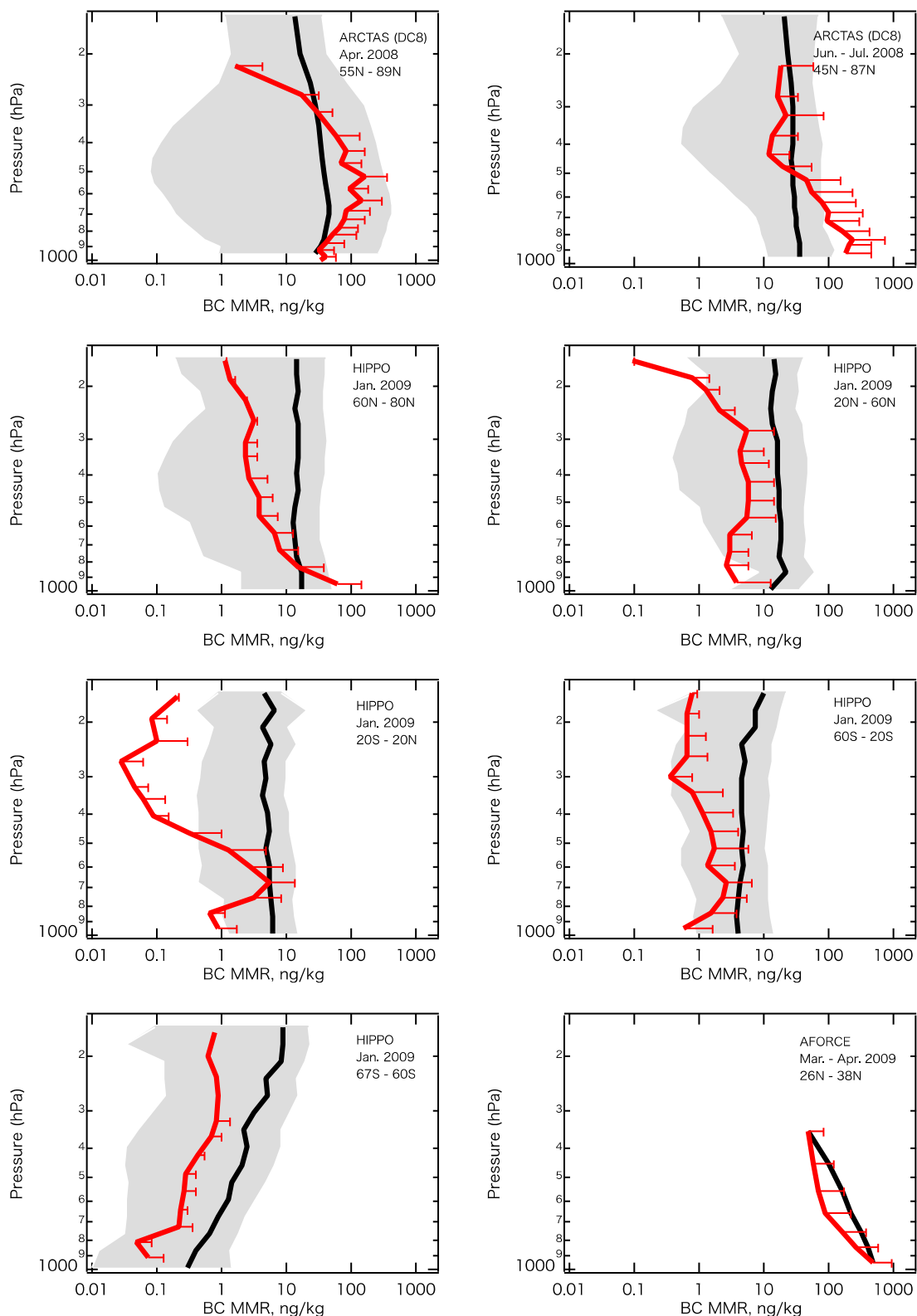
3

Figure 7.9: Bar chart plots summarizing the annual, seasonal or monthly mean mass concentration ($\mu\text{g m}^{-3}$) of six major types of aerosol particles in diameter smaller than $10 \mu\text{m}$ with at least an entire year data from various rural and urban sites in nine continental areas of the world. These include: 1) rural U. S. (Chow et al., 1993; Liu et al., 2005a; Malm and Schichtel 2004; Malm et al., 1994); urban U. S. (Chow et al., 1993; Ito et al., 2004; Kim et al., 2000; Liu et al., 2005a; Malm and Schichtel 2004; Sawant et al., 2004); 2) South America (Artaxo et al., 1998; Artaxo et al., 2002; Bourotte et al., 2007; Celis et al., 2004; Fuzzi et al., 2007; Gioda et al., 2011; Mariani and Mello 2007; Martin et al., 2010; Morales et al., 1998; Souza et al., 2010); 3) rural Europe (Gullu et al., 2000; Hueglin et al., 2005; Kocak et al., 2007; Putaud et al., 2004; Puxbaum et al., 2004; Querol et al., 2001; Querol et al., 2009; Querol et al., 2004; Rodriguez et al., 2002; Rodriguez et al., 2004; Salvador et al., 2007; Theodosi et al., 2010; Viana et al., 2008; Yin and Harrison 2008; Yttri 2007); urban Europe (Hueglin et al., 2005; Lenschow et al., 2001; Lodhi et al., 2009; Lonati et al., 2005; Perez et al., 2008; Putaud et al., 2004; Querol et al., 2001; Querol et al., 2006; Querol et al., 2004; Querol et al., 2008; Rodriguez et al., 2002; Rodriguez et al., 2004; Roosli et al., 2001; Viana et al., 2007; Viana et al., 2006; Yin and Harrison 2008); 4) rural Africa (Maenhaut et al., 1996; Mkoma 2008; Mkoma et al., 2009a; Mkoma et al., 2009b; Nyanganyura et al., 2007; Weinstein et al., 2010); urban Africa (Favez et al., 2008; Mkoma 2008; Mkoma et al., 2009a); 5) high Asia, with altitude larger than 1680 m. (Carrico et al., 2003; Decesari et al., 2010; Ming et al., 2007a; Qu et al., 2008; Ram et al., 2010; Rastogi and Sarin 2005; Rengarajan et al., 2007; Shresth et al., 2000; Zhang et al., 2001; Zhang et al., 2008; Zhang et al., 2011a); 6) rural China (Hagler et al., 2006; Hu et al., 2002; Zhang et al., 2011a); urban China (Cheng et al., 2000; Hagler et al., 2006; Oanh et al., 2006; Wang et al., 2003; Wang et al., 2005b; Wang et al., 2006; Xiao and Liu 2004; Yao et al., 2002; Ye et al., 2003; Zhang et al., 2002; Zhang et al., 2011a; Zhang et al., 2011b); 7) South-East and East Asia (Han et al., 2008; Khan et al., 2010; Kim et al., 2007; Lee and Kang 2001; Oanh et al., 2006); 8) urban South Asia (Chakraborty and Gupta 2010; Khare and Baruah 2010; Kumar et al., 2007; Lodhi et al., 2009; Raman et al., 2010; Rastogi and Sarin 2005; Safai et al., 2010; Stone et al., 2010); 9) urban Oceania (Chan et al., 1997; Maenhaut et al., 2000; Radhi et al., 2010; Wang and Shooter 2001; Wang et al., 2005a).

27

28

1



2

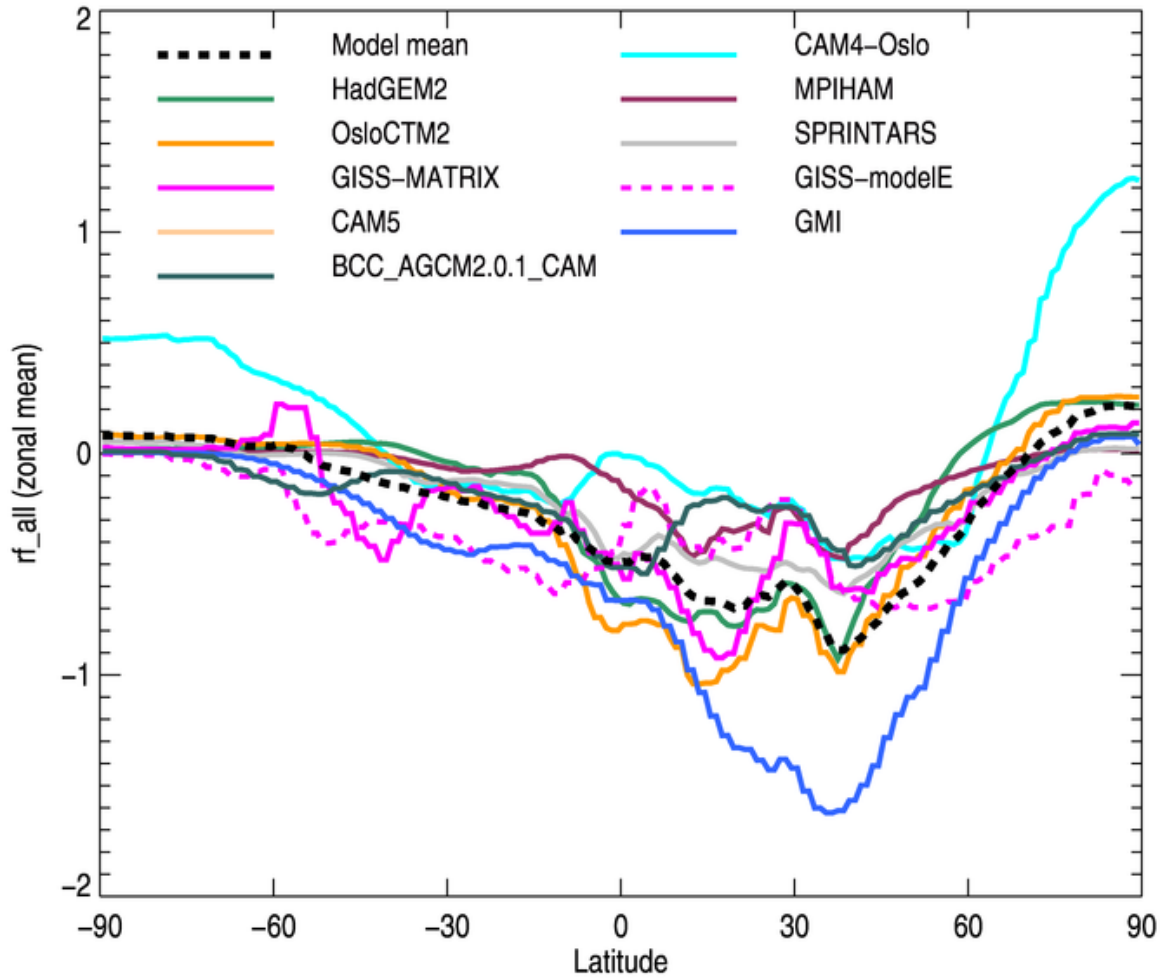
3

4 **Figure 7.10:** Comparison of BC profiles as measured during the ARCTAS, HIPPO1 and FORCE-A campaigns and
 5 simulated by a range of global aerosol models. [PLACEHOLDER FOR SECOND ORDER DRAFT: will be updated
 6 from AeroCom and CMIP5 models]

7

8

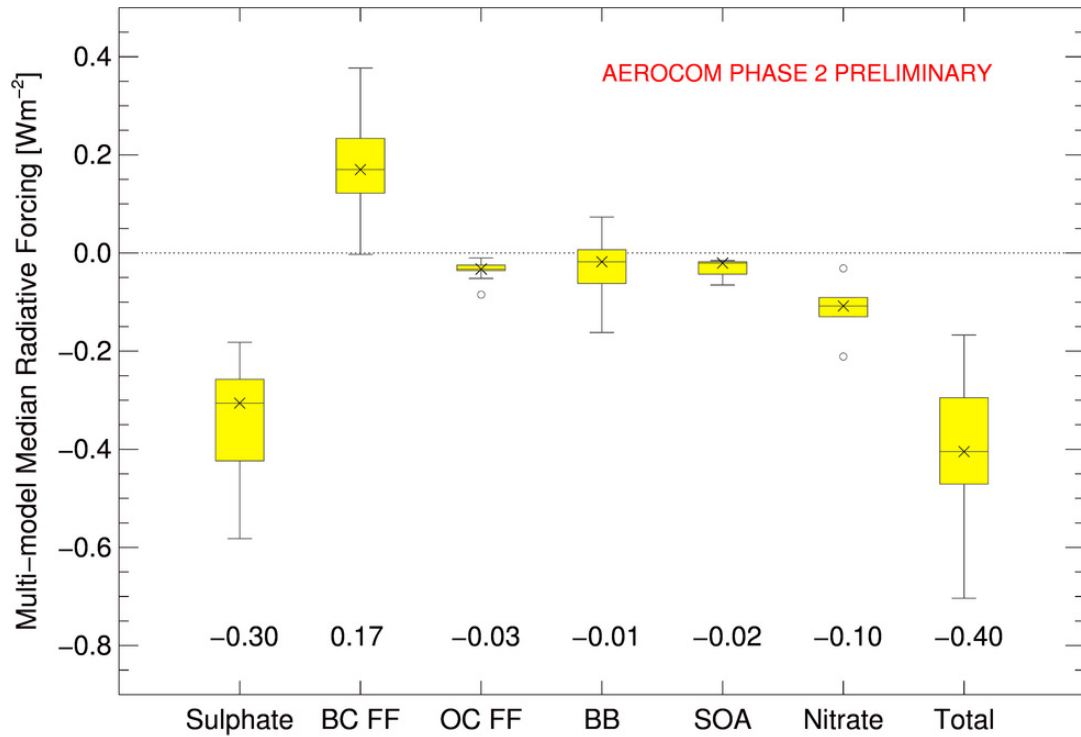
1



2
3
4
5
6
7

Figure 7.11: Zonal mean total aerosol direct radiative forcing from the different AeroCom models. No adjustment for missing species has been applied.

1



2

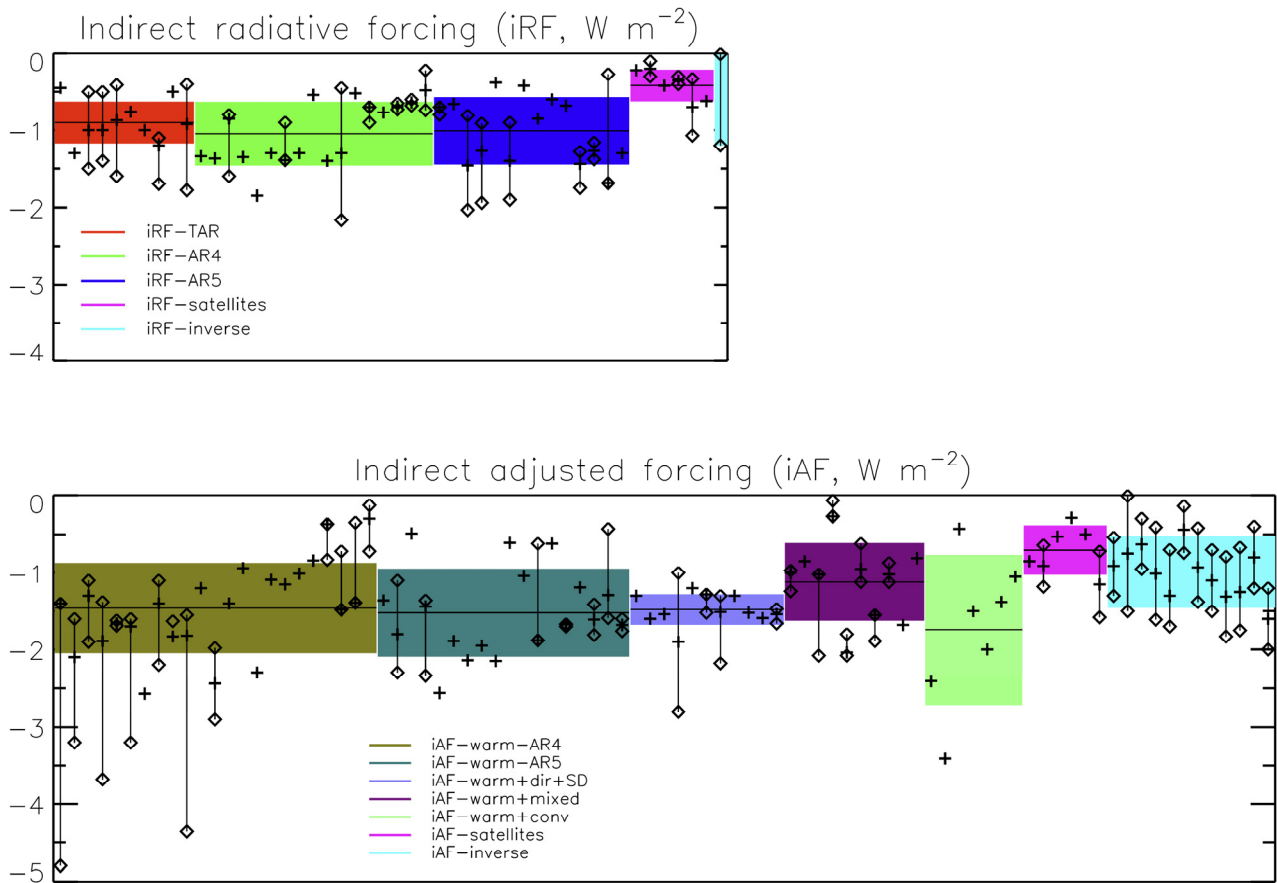
3

4 **Figure 7.12:** Median, full range and 5%–95% range of AeroCom model direct radiative forcing by species and the total
 5 direct forcing. The total direct forcing has been adjusted to take account of missing species in some models by adding
 6 the median value of the species forcing from the remaining models.

7

8

1



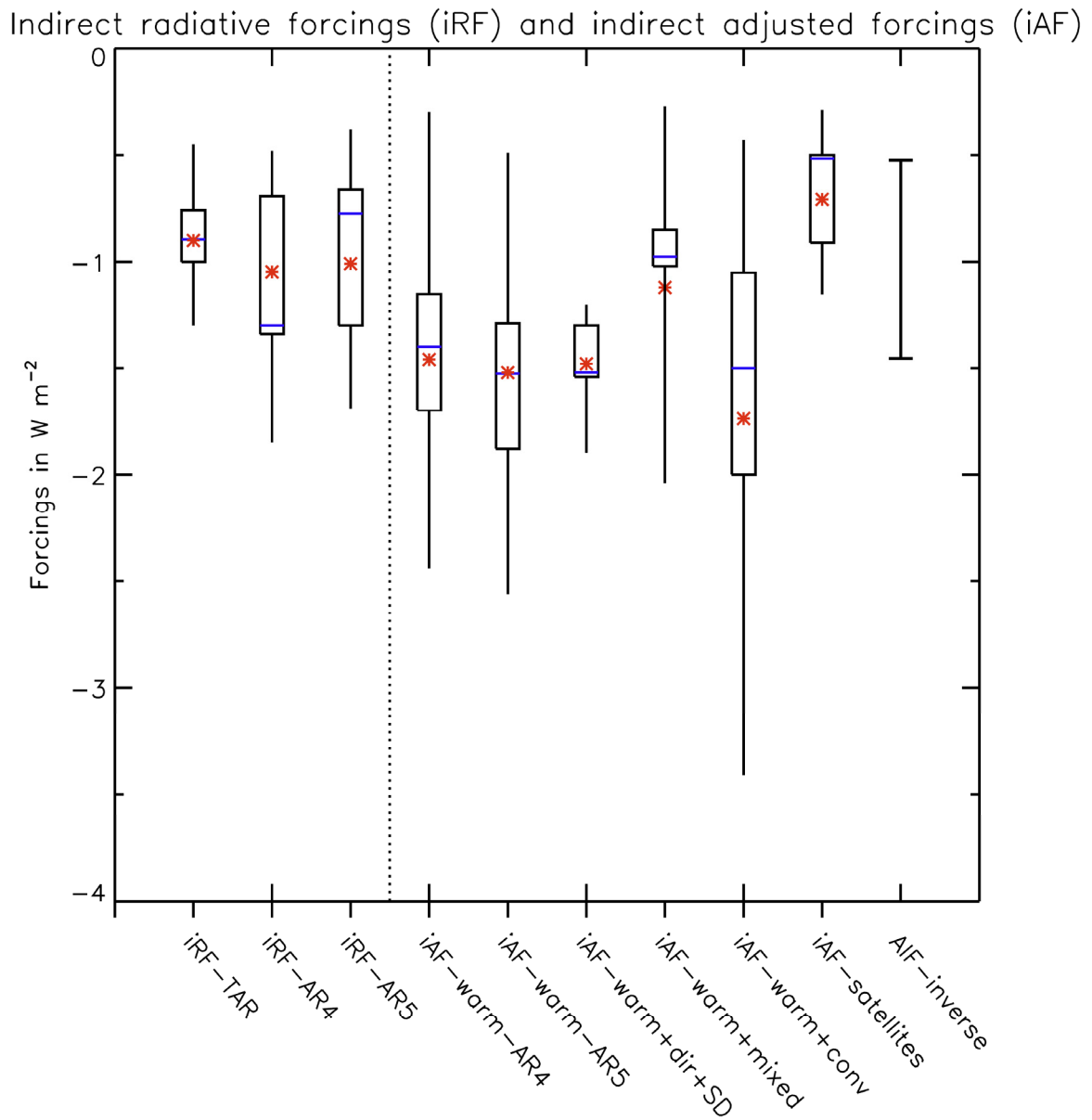
2

3

Figure 7.13a: Model, satellite and inverse estimates of the iRF and the iAF since 1993. For each paper the best estimate per model is shown as a plus-sign. If multiple estimates or uncertainties are given in a paper, the ranges are shown as vertical lines bounded by diamonds. The thin horizontal lines denote the average of the respective group and the width of the coloured box denotes its standard deviation. The iRF studies from GCMs are divided into those published prior to TAR: iRF-TAR (Boucher and Lohmann 1995; Chuang et al., 1997; Feichter et al., 1997; Jones et al., 1994; Kaufman and Chou 1993; Kiehl et al., 2000; Lohmann and Feichter 1997; Lohmann et al., 2000; Rotstayn 1999), between TAR and AR4: iRF-AR4 (Chen and Penner 2005; Chuang et al., 2002; Ghan et al., 2001; Hansen et al., 2005; Jones et al., 2001; Kristjansson 2002; Ming et al., 2005; Penner et al., 2006; Quaas and Boucher 2005; Quaas et al., 2004; Rotstayn and Penner 2001; Rotstayn and Liu 2003; Suzuki et al., 2004; Takemura et al., 2005; Williams et al., 2001) and since 2007: iRF-AR5 (Barahona et al., 2011; Bellouin et al., 2011; Haerter et al., 2009; Kvalevåg and Myhre 2007; Lohmann et al., 2007; Lohmann et al., 2010; Penner et al., 2011; Rotstayn and Liu 2009; Storelvmo 2011; Storelvmo et al., 2009; Wang and Penner 2009). iAF studies on liquid clouds that include the cloud albedo and cloud lifetime effect are also divided into those published until 2006: iAF-liquid-AR4 (Easter et al., 2004; Ghan et al., 2001; Johns et al., 2006; Jones et al., 2001; Kristjansson 2002; Kristjansson et al., 2005; Lohmann 2002b; Lohmann and Feichter 1997; Lohmann et al., 2000; Menon et al., 2002; Ming et al., 2005; Peng and Lohmann 2003; Penner et al., 2003; Penner et al., 2006; Quaas et al., 2006; Rotstayn 1999; Rotstayn and Penner 2001; Rotstayn and Liu 2005; Storelvmo et al., 2006; Takemura et al., 2005; Williams et al., 2001) and since 2007: iAF-liquid-AR5 (Chen et al., 2010; Ghan et al., 2011b; Hoose et al., 2009; Kirkevåg et al., 2008; Makkonen et al., 2011; Menon and DelGenio 2007; Ming et al., 2007b; Penner et al., 2011; Quaas et al., 2009b; Rotstayn and Liu 2009; Storelvmo et al., 2008a); iRF and iAF estimates that involve satellite data are shown in pink: iRF-satellites (Dufresne et al., 2005; Lebsack et al., 2008; Quaas and Boucher 2005; Quaas et al., 2008; Quaas et al., 2009b; Storelvmo et al., 2009) and iAF-satellites (Lohmann and Lesins 2002; Quaas et al., 2006; Quaas et al., 2009b; Sekiguchi et al., 2003), inverse estimates for the iRF and iAF are shown in turquoise: iRF-inverse (Knutti et al., 2002) and iAF-inverse (Anderson et al., 2003; Andronova and Schlesinger 2001; Church et al., 2011; Forest et al., 2006; Forest et al., 2002; Gregory et al., 2002; Hansen et al., 2011; Harvey and Kaufmann 2002; Huber and Knutti 2011; Libardoni and Forest 2011; Murphy et al., 2009; Shindell and Faluvegi 2009; Stott et al., 2006) in turquoise. iAF studies that include the direct and semi-direct effect in lilac: iAF-liquid+dir+SD (Ghan et al., 2011a; Lohmann and Feichter 2001; Lohmann et al., 2007; Posselt and Lohmann 2008; Posselt and Lohmann 2009; Quaas et al., 2004; Quaas et al., 2006; Quaas et al., 2009b; Rotstayn et al., 2007; Salzmann et al., 2010), those that additionally consider aerosol effects on mixed-phase clouds in purple: iAF-liquid+mixed (Hoose et al., 2008; Hoose et al., 2010b; Jacobson 2006; Lohmann 2004; Lohmann and Diehl 2006; Lohmann and Hoose 2009; Lohmann and Ferrachat 2010; Salzmann et al., 2010; Storelvmo et al., 2008a; Storelvmo et al., 2008b), and those that

1 treat aerosol effects in stratiform and convective clouds in green: iAF-liquid+conv (Koch et al., 2009a; Lohmann 2008;
2 Menon and Rotstayn 2006; Menon and DelGenio 2007; Unger et al., 2009; Wang et al., 2011b). For the inverse
3 estimates no best estimate is shown and the turquoise colour box denotes the average of the lower and upper bounds of
4 these studies, respectively.
5
6

1



2

3

4

5

6

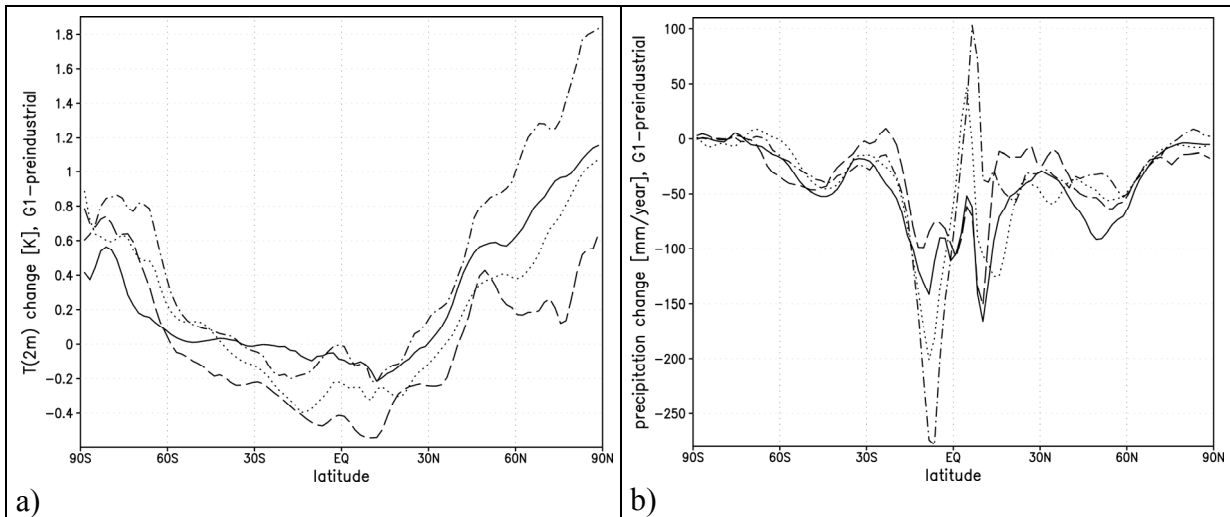
7

8

9

Figure 7.13b: Box plots of model, satellite and inverse estimates of the IF and the AIF since 1993 for the same groups of estimates as in Figure 7.13a provided at least 6 estimates are available. Displayed are the averages (red stars), median values (blue lines), 33% and 67% percentiles (box boundaries) and 5% and 95% percentiles (ends of vertical lines) except for the inverse estimates, which is an expert assessment of the combined estimate of multiple inverse estimates.

1



2

3

4

Figure 7.14: Multi-model mean of the residual surface temperature and precipitation changes from GeoMIP simulations with a simultaneous fourfold increase in CO₂ and a reduction in solar forcing which has been adjusted in each model to maintain the top of atmosphere net flux imbalance within $\pm 0.1 \text{ W m}^{-2}$ (Kravitz et al., 2011).

5

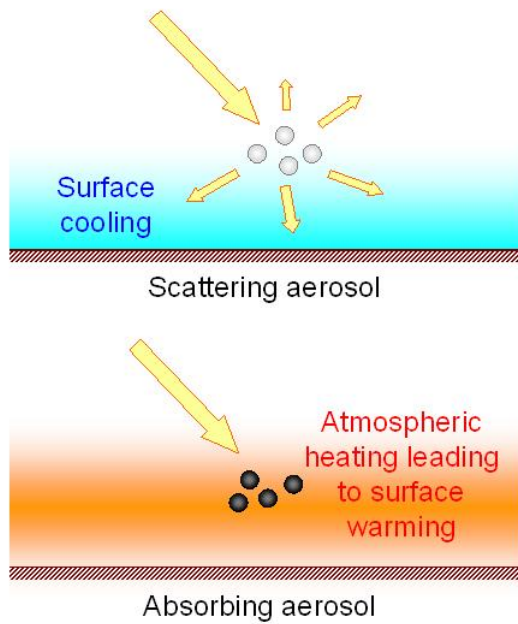
6

7

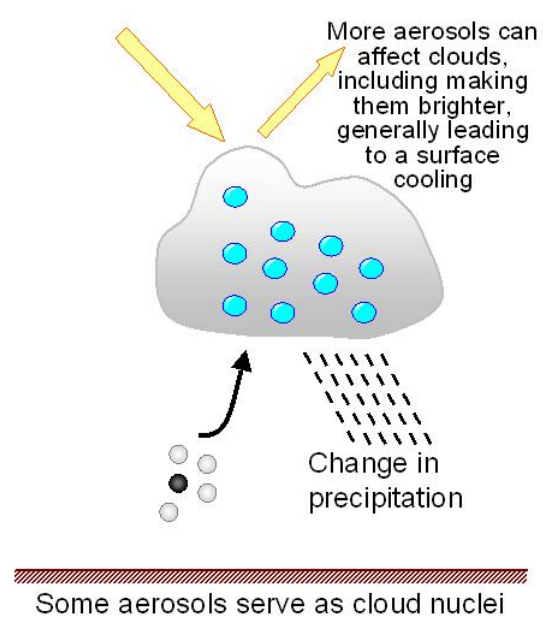
8

1

Aerosol direct and semi-direct effects



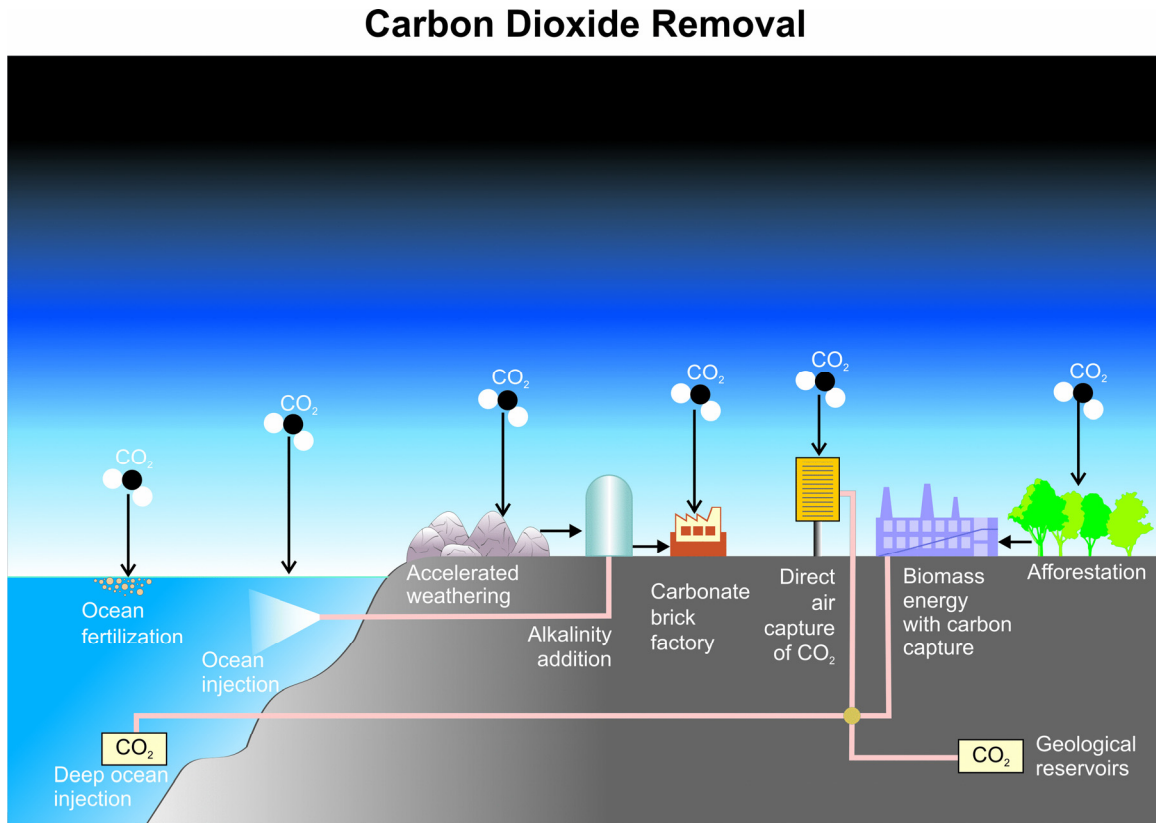
Aerosol indirect effects



2
3
4
5
6

FAQ 7.1, Figure 1: Overview of aerosol direct and indirect effects on climate.

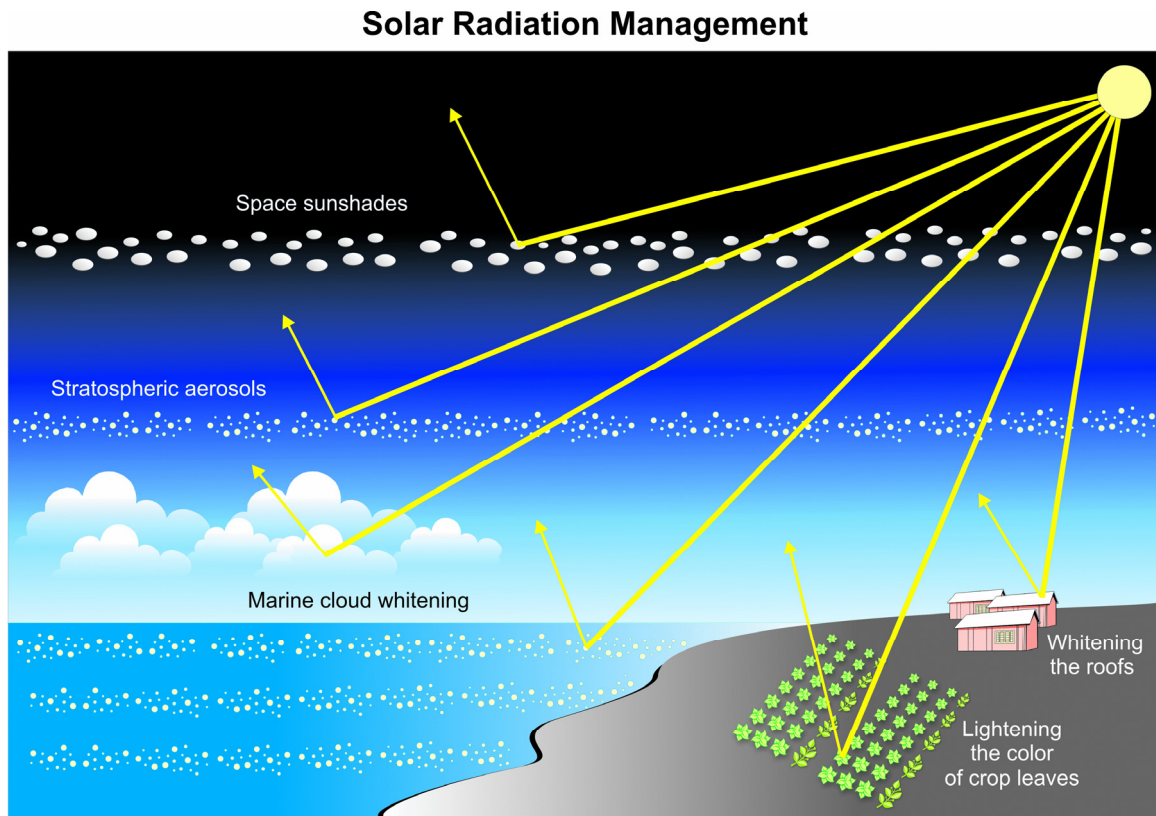
1



2
3
4
5
6

FAQ 7.3, Figure 1: Overview of carbon dioxide removal methods.

1



2

3

4

5

FAQ 7.3, Figure 2: Overview of solar radiation management methods.