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Review

Possible impacts of anthropogenic and natural aerosols on Australian climate: a review

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ABSTRACT: A review is presented of the aerosol–climate interaction with specific focus on the Australian region. The uncertainties associated with this interaction are much larger than those associated with greenhouse gases or other forcing agents, and are currently a major obstacle in climate-change research. However, new research suggests that aerosol effects are of comparable importance to greenhouse gases as a driver of recent climate trends in the Southern Hemisphere, including Australia. A significant new result from climate modelling is that anthropogenic aerosol over Asia affects meridional temperature gradients and atmospheric circulation, and may have caused an increase in rainfall over north-western Australia. Global ocean circulation provides another mechanism whereby aerosol changes in the Northern Hemisphere can affect climate in the Southern Hemisphere, suggesting an urgent need for further targeted studies using coupled ocean-atmosphere global climate models. To better model climate variability and climate change in the Australian region, more research is needed into the sources of aerosol and their precursors, their atmospheric distributions and transformations, and how to incorporate these processes robustly in global climate models (GCMs). The following priorities are suggested for further research in Australia linking aerosol observations and modelling: natural aerosol over the Southern Ocean, tropical biomass-burning aerosol in Indonesia and Australia, secondary organic aerosol (SOA) from volatile organic compounds (VOCs), wind-blown dust and modulation of rainfall by anthropogenic aerosol. Copyright © 2008 Royal Meteorological Society

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1. Introduction

Aerosol effects are the most uncertain of the radiative forcing components by which human activity is altering our climate, according to the Intergovernmental Panel on Climate Change (IPCC); Forster *et al*., 2007). There are three ways in which aerosols are known to affect climate. The direct effect of aerosol on short and long wave radiation comes from the scattering and absorption properties of the aerosol. The direct effect is uncertain, with an estimated net global-mean forcing of −0.1 to −0.9 W m^{-2}. The indirect effects of aerosol on climate occur when aerosols modify cloud properties by acting as cloud condensation nuclei (CCN). An increase in aerosol concentration causes the liquid water in clouds to be distributed over a larger number of smaller cloud droplets, leading to an increase in cloud albedo (Twomey, 1974). This first indirect effect is now considered to be well established, although its magnitude is very uncertain, with a range of −0.3 to −1.8 W m^{-2} for its global-mean radiative forcing, according to the IPCC. The second indirect effect, whereby smaller cloud droplets cause a decrease in the precipitation efficiency, is even more uncertain, and the IPCC does not give an estimate of its magnitude. These uncertainty ranges are much larger than those associated with greenhouse gases or other forcing agents, and are currently a major obstacle in climate-change research.

The effect of atmospheric aerosol on climate has been observed empirically for some time. After the great eruption of Tambora in 1815, the Northern Hemisphere had a 'year without a summer' (Stommel and Stommel, 1983) and more recently the effects of major volcanic eruptions on global temperatures have been observed (e.g. Soden *et al*., 2002). There is also palaeoclimatic evidence

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for an El Niño-like response to volcanic eruptions, at least in a statistically averaged sense (Adams *et al.*, 2003). On inter-annual timescales, there is evidence that Saharan dust modulates the formation of tropical cyclones over the North Atlantic (Evan *et al.*, 2006; Lau and Kim, 2007a,b).

It has been understood for over a decade that aerosol pollution produced by human activity partly offsets the warming effect of greenhouse gases. The inclusion of anthropogenic aerosols in climate models is necessary to obtain a realistic simulation of historical global-mean temperature changes (e.g. Mitchell *et al.*, 1995). Recent studies have suggested large climatic effects due to aerosols in the Northern Hemisphere, primarily due to strong spatial variations in the distribution of aerosols. This spatial inhomogeneity can lead to substantial changes in atmospheric circulation and rainfall, especially in the tropics, where circulation is known to be sensitive to large-scale horizontal temperature gradients (Rotstayn and Lohmann, 2002a). The best-known example of this temperature gradient-circulation connection is the El Niño Southern Oscillation (ENSO), whereby changes in sea-surface temperatures (SSTs) in the eastern tropical Pacific affect circulation and rainfall over much of the globe. Possible impacts of anthropogenic aerosols on weather and climate include summertime floods and droughts in China (Menon *et al.*, 2002), droughts in the Sahel (Rotstayn and Lohmann, 2002a), and a weakening of the South Asian Monsoon (Ramanathan *et al.*, 2005). Collectively, these studies suggest large impacts of anthropogenic aerosols on the climate of the Northern Hemisphere.

Anthropogenic aerosol concentrations are much lower in the Southern Hemisphere than in the Northern Hemisphere. Until recently, evidence of anthropogenic aerosol effects on the climate of the Southern Hemisphere has been confined to those associated with biomass burning in the tropics (e.g. Keil and Haywood, 2003). However, recent results from the CSIRO climate model (Cai *et al.*, 2006, 2007; Cai and Cowan, 2007; Rotstayn *et al.*, 2007) suggest that anthropogenic aerosols (principally from the Northern Hemisphere) are important for understanding late twentieth century trends in the climate of the Southern Hemisphere, including Australia. Other studies have suggested the possibility of climatic or meteorological effects due to aerosol in the Australian region (e.g. Davison *et al.*, 2004; Mitchell *et al.*, 2006).

The aim of this article is to summarize the state of knowledge of the aerosol–climate connection and describe the possible impacts of aerosols on the climate of the Australian region. In Section 2 of this article, basic aerosol concepts are briefly reviewed. In Section 3, the treatment of aerosols in the CSIRO climate model is described and critically evaluated. In Section 4, recent simulations with the CSIRO climate model, which suggest substantial effects of aerosol on Australian and Southern Hemisphere climate, are reviewed. In Section 5, possible climatic effects due to Australian-region aerosol are considered. In Section 6, key research directions are proposed.

2. Aerosol fundamentals

Aerosols can be defined as an assembly of solid and liquid particles suspended in a gaseous medium long enough to be observed (Baron and Willeke, 2001). Atmospheric particles range in size from around 0.001 to 100 μm in diameter and their composition and size are dependent on the source and mechanism of formation and subsequent chemical and physical reactions of the particles within the atmosphere. Particles can be directly emitted into the atmosphere (as primary particles) or be formed in the atmosphere by gas-to-particle conversion (as secondary particles).

Typically, the aerosol size distribution is comprised of four modes, referred to as the coarse-particle (2.5–100 μm in diameter), accumulation (0.1–2.5 μm), Aitken (0.01–0.1 μm) and nucleation modes (less than 10 nm) (Finlayson-Pitts and Pitts, 2000). Particles that have the greatest impact on climate reside in the accumulation mode. Their size and composition (typically sulfates and organic carbon compounds) mean that they have longer lifetimes in the atmosphere than other particles, typically 3–10 days and hence the most significant effect on visibility, cloud formation and atmospheric chemistry. In the presence of large concentrations of dust, the coarse-particle mode can also be optically active and hence be climatically important. Most aerosol–climate-related processes are not directly mass-concentration dependent, for example, light scattering is more closely related to particle area, and cloud-nucleating capacity to particle number, in both cases above a particular size threshold. The important diameter range is relatively small, falling between about 0.05 and 1 μm (roughly the aerosol accumulation mode).

The degree of chemical mixing in particles influences the optical and hygroscopic properties of the aerosol. The chemical composition determines the refractive index of the particle; for example, particles comprised only of sulfates and organics predominantly scatter solar energy, whereas particles containing black carbon (BC), iron oxide or some organic acids also strongly absorb solar energy. Together the scattering and absorption coefficients define the extinction coefficient and single scattering albedo and these are key coefficients for modelling the aerosol–radiation interaction. The efficiency of the scattering is highly dependent on particle size, with the highest relative efficiency occurring when particle diameter is similar to the wavelength (~0.1–1 μm). Absorption efficiency, however, is less dependent on particle size, and more dependent on the presence of solar energy–absorbing species such as elemental carbon; typically this resides in particles less than 0.1 μm (Seinfeld and Pandis, 1998).

The presence of soluble components such as sulfate, nitrate and soluble organics governs the hygroscopicity

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of the aerosol, and thus influences scattering and the activation of aerosols to form cloud droplets. In the absence of aerosols, cloud droplets cannot form without supersaturations of several hundred percent, which do not occur in the atmosphere. Soluble aerosols allow the formation of cloud droplets at supersaturations of less than 1%, because a soluble particle reduces the equilibrium vapour pressure over a droplet containing that particle. This explains the vital role of aerosols as CCN, without which clouds could not form under realistic conditions.

The atmosphere over Australia is influenced by a number of aerosol sources, including soil-dust, sea-salt, biomass burning and the products of gas-to-particle conversion [secondary organic aerosol (SOA) and secondary sulfate]. The major distributed anthropogenic sources are the major capital cities in each state, although there are several significant, isolated point sources, such as power stations (e.g. La Trobe Valley in Victoria and Hunter Valley in NSW) and ore-processing facilities (e.g. Gladstone in Queensland, Mount Isa in Queensland and Port Pirie in South Australia). While rough estimates of the contribution of these sources to the mass of aerosol on a global scale have been made (Galbally and Freney, 1978; Kiehl and Rhode, 1995; Satheesh and Moorthy, 2005; Goldstein and Galbally, 2007), the contribution different sources make to the climatically important accumulation mode aerosol have yet to be accurately determined. Goldstein and Galbally (2007) suggested that biogenic SOA may be a major type of aerosol that is seriously underestimated in global inventories. There has been minimal study of biogenic SOA over Australia.

The interaction between aerosol and climate is complex and bi-directional, with inevitable blurring between some natural and anthropogenic sources. For example, aerosol from biomass burning and SOA have a complex relationship with inter-annual rainfall variations. Most natural aerosol sources are strongly dependent on meteorological factors as are the removal processes for all aerosols. Many of these processes are non-linear. Hence, understanding such linkages is important for accurate prediction of the overall aerosol impact and possibly for understanding inter-annual and longer term natural climate variability. Figure 1 shows important aerosol processes that influence climate and must be accurately represented in future generations of climate models.

The Fourth Assessment Report of the IPCC estimates the global-mean net radiative forcing due to anthropogenic aerosol as -1.2 W m^{-2} for the period 1750–2005, with a large uncertainty (Forster *et al.*, 2007). This has almost half the magnitude of the forcing due to long-lived greenhouse gases (2.6 W m^{-2}), which indicates that anthropogenic aerosols have substantial climatic effects. The potency of aerosol as a climate-forcing agent is further increased by the spatial variations referred to in the Introduction. Also, aerosols with an absorbing component (such as dust or aerosol from biomass burning) have a relatively small effect on the radiative flux at the top of the atmosphere, and a much larger effect on surface radiation and atmospheric heating. Partially absorbing aerosols cause surface cooling and lower atmospheric heating; this tends to increase atmospheric stability and reduce convection (Satheesh and Moorthy, 2005). The usefulness of the top-of-atmosphere radiative forcing as a measure of climatic importance thus breaks down for partially absorbing aerosol (Forster *et al.*, 2007). Also, the radiative forcing concept does not measure the role of natural aerosol and its interaction with climate variability and climate change. Examples of this are given in Section 5.

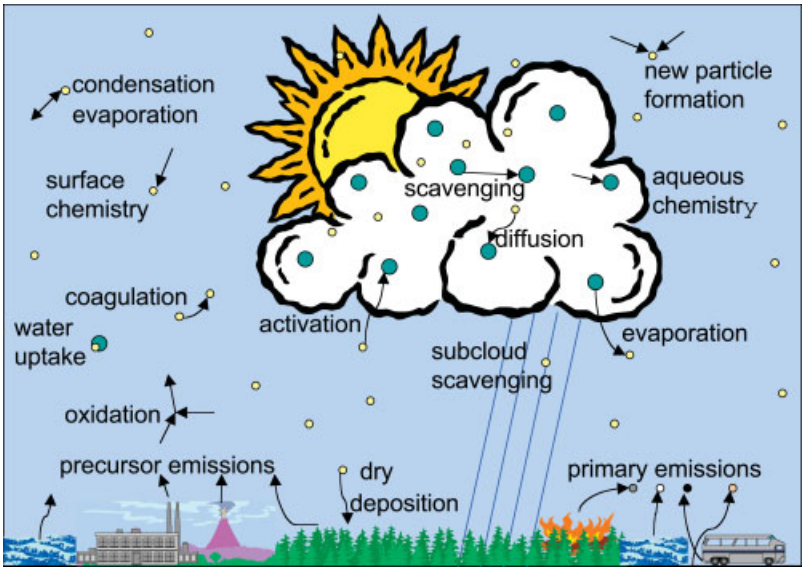

Figure 1. Important aerosol processes that influence climate and must be accurately represented in future generations of climate models. Reproduced from Ghan and Schwartz (2007) by permission of the American Meteorological Society. This figure is available in colour online at www.interscience.wiley.com/ijoc

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3. Treatment of aerosol in the CSIRO climate model

3.1. Description of the treatment and the problem of simplification

Modelling of aerosols in a global climate model (GCM) is highly challenging and uncertain, and many simplifications are necessary. An interactive aerosol treatment was implemented in the CSIRO GCM (Rotstayn and Lohmann, 2002b; Rotstayn *et al*., 2007, hereafter R2007) and its strengths and limitations were discussed by R2007. This aerosol treatment is comparable to some of the more detailed treatments in the GCMs that contributed to the IPCC's Fourth Assessment. More advanced aerosol treatments for global models have been developed (e.g. Liu *et al*., 2005; Spracklen *et al*., 2007) and are gradually being introduced into GCMs. The treatments used in GCMs generally lag those in the best global aerosol models by a few years, in part due to the large computational requirements of more detailed models. Although we describe the aerosol treatment in the CSIRO GCM as simplified, the version of the model with aerosols takes roughly twice as long to run as the version without aerosols, which explains why such simplified treatments are necessary.

The aerosol species treated interactively in the CSIRO GCM are sulfate, organic aerosol (OA), BC, mineral dust and sea-salt. All of the above aerosol types, except for sea-salt, are treated prognostically by the model, so that equations are included for their emission, transformation, transport and removal at each time step. In all, there are 11 prognostic variables in the aerosol scheme: dimethyl sulfide (DMS), sulfur dioxide (SO_2), sulfate, hydrophobic and hydrophilic forms of BC and OA and four different sizes of mineral dust. In addition, two modes of sea-salt are diagnosed as a function of wind speed in the marine boundary layer, but are not treated prognostically.

The model includes both anthropogenic and natural sources of aerosol and aerosol precursors. Historical anthropogenic emissions are included for sulfur (Smith *et al*., 2004) and carbonaceous aerosols (Ito and Penner, 2005). The sulfur emissions occur mostly as SO_2 (which is oxidized to sulfate in the atmosphere), with just 3% as primary sulfate aerosol. The carbonaceous aerosol emissions include primary sources of BC and OA from the burning of fossil fuel, open vegetation and biofuel. Since secondary sources of anthropogenic OA were not included in the inventory, the fossil-fuel OA emissions were scaled up as a simple way to allow for secondary sources of OA (see R2007). Natural sources of sulfur are SO_2 from non-eruptive volcanoes and biogenic emissions of DMS from oceans. For natural SOA from vegetation (Guenther *et al*., 1995), a yield of 13% is assumed for rapid conversion of monoterpenes to OA, giving an annual source of 16.4 Tg C. The treatment of mineral-dust emission (Ginoux *et al*., 2001) is based on satellite analyses that identified major dust sources as topographic depressions in which a sufficiently deep layer of alluvium was able to accumulate.

Aerosol processes treated in the model are transport, chemical transformation, aerosol removal by wet and dry deposition, aerosol-radiative effects and cloud-droplet nucleation. Transport of aerosols and precursor gases occurs by advection, vertical turbulent mixing and vertical transport inside deep convective clouds. The model has a rudimentary treatment of the tropospheric sulfur cycle, which includes oxidation of DMS to SO_2 by gas-phase reactions, and oxidation of SO_2 to sulfate by in-cloud and gas-phase reactions. The chemical transformation of carbonaceous aerosol is poorly understood, so it is treated even more crudely: hydrophobic forms of BC and OA are assumed to transform into their hydrophilic forms with a timescale of 1.15 days. Wet deposition (scavenging) of aerosol occurs both in clouds and below clouds, by rain and snow. Dry deposition to the surface is treated via deposition velocities that depend on the aerosol species and surface type. The model includes effects of the above aerosol types on short-wave radiation, using prescribed size distributions and the assumption that all aerosol species exist as external mixtures (except for BC and OA, which are internally mixed). Cloud-droplet nucleation is a complex process that is difficult to resolve in a GCM, so the cloud-droplet number concentration in stratiform clouds is parameterized as an empirical function of the concentration of sulfate, hydrophilic OA and sea-salt. Cloud-droplet number concentration affects cloud albedo and precipitation efficiency, so it has an important role in the model's simulation of the first and second indirect effects (Rotstayn and Liu, 2003, 2005).

The treatment of every process mentioned above is over-simplified or highly uncertain, and it is beyond the scope of this article to discuss them in detail. One important limitation of the model is that the prognostic aerosol schemes are 'bulk' treatments, meaning that only the mass of each aerosol species is predicted, and the number concentration is assumed proportional to the mass concentration. This implies that new particle formation, growth of particles and interactions between particles cannot be treated. Further, some additional aerosol types, such as nitrate, are not included in the model, although they are thought to exert substantial climatic effects (Adams *et al*., 2001). Emission inventories are also highly uncertain, even for the present climate. A more detailed assessment of the model's design and its limitations was given by R2007.

3.2. Evaluation of the aerosol simulation

R2007 compared modelled distributions of mid-visible aerosol optical depth (AOD) with a satellite-based composite compiled by Kinne *et al*. (2006), which suggested that the model tends to underestimate AOD. However, the passive satellite retrievals used by Kinne *et al*. are prone to overestimate AOD (due to contamination by high clouds), and this can exaggerate the impression of an underestimate in the GCM. For a complementary view, it is useful to compare the modelled clear-sky mid-visible AOD with another new climatology, derived by Kinne

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(2008). This climatology prioritizes high-quality measurements from the surface-based Aerosol Robotic Network (AERONET) (Holben *et al*., 2001), but falls back onto the median output fields from the AeroCom models (Kinne *et al*., 2006) if AERONET data are unavailable or considered too poor in quality. Thus, the climatology can be called an 'AERONET–AeroCom blend'. Figure 2 confirms, for the June–August (JJA) season, the broad finding that this version of the model underestimates AOD for the modern climate. As discussed by R2007, the underestimate in areas strongly affected by anthropogenic aerosol is substantially due to underestimated emissions of carbonaceous aerosols in the historical inventory. Emissions of carbonaceous aerosols are generally considered to be among the major uncertainties in global aerosol modelling (e.g. Liu *et al*., 2005). Over remote oceans, the underestimate of AOD may be due to the simple treatment of sea-salt aerosol, which assumes that sea-salt is well mixed in the marine boundary layer, but has zero concentration above the boundary layer. However, another possible explanation is that the global production of SOA from natural sources is seriously underestimated in the CSIRO GCM. The model's source of natural SOA (16 Tg C per annum, which is typical of current models) was suggested to be a serious underestimate by Heald *et al*. (2005). It is also much smaller than the recent estimates from Goldstein and Galbally (2007), which exceed 300 Tg C per annum.

In a regional context, the models underestimate AOD over tropical Australia and Indonesia, which are affected by biomass burning in JJA. The main cause of the problem seems to be an underestimate of these biomass-burning emissions in the historical inventory (Ito and Penner, 2005) used by the model. Thus, this version of the model is probably not an effective tool to evaluate any possible impacts of these aerosols on Australian

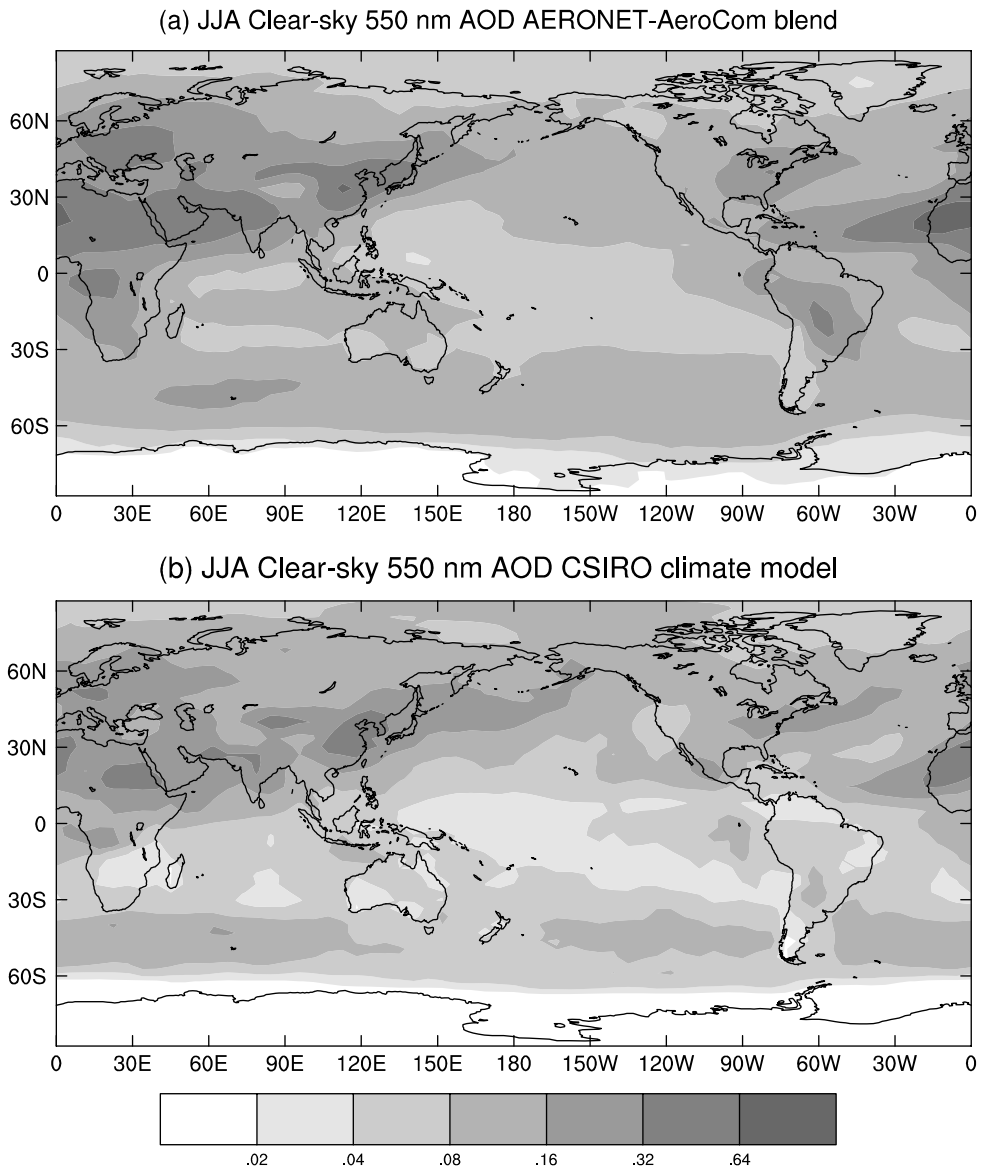

Figure 2. Clear-sky aerosol optical depth at 550 nm for June–August from (a) the climatology (AERONET-AeroCom blend), and (b) the CSIRO GCM, using emissions for the year 2000. Reproduced from Rotstayn (2007).

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climate. Despite this, the model successfully captures the dominance of aerosol in the Northern Hemisphere relative to that in the Southern Hemisphere, and the large source over Asia. The implications of these important points are considered in the next section.

R2007 estimated the net (direct plus indirect) global-mean aerosol-radiative forcing in the CSIRO GCM as -1.2 W m^{-2} for the period 1870–1990, which agrees well with the best estimate of -1.2 W m^{-2} from the IPCC's Fourth Assessment (Forster *et al.*, 2007). This gives some confidence that the CSIRO GCM does not seriously overestimate the global effects of aerosols.

4. Simulated effects of global aerosol on Australian climate

One of the most significant recent results from GCM modelling of Australian climate is the finding that Asian anthropogenic aerosol affects the temperature gradient and monsoonal winds between Asia and Australia, and may have caused an increase in rainfall over north-western Australia.

R2007 used a low-resolution version of the Mk3 CSIRO GCM to perform ensembles of coupled ocean–atmosphere simulations for the period 1871–2000, with and without the effects of aerosols. The model is based on an earlier high-resolution version of the Mk3 atmospheric GCM (Gordon *et al.*, 2002), which was upgraded by inclusion of the aerosol and radiation treatments described above. Along with the direct and indirect aerosol effects described above, the model was forced by changes in long-lived greenhouse gases, ozone, volcanic eruptions and solar variations. An eight-run ensemble with all of these forcings (ALL ensemble) and a further eight runs with all forcings except those related to anthropogenic aerosols (AXA ensemble) were performed. The AXA ensemble only differs from the ALL ensemble in that the anthropogenic emissions of sulfur, OA and BC were fixed at their 1870 levels. To isolate the effects of Asian aerosols, another eight-run ensemble was performed, identical to the AXA ensemble, except that Asian anthropogenic emissions of sulfur, OA and BC were allowed to vary with time. R2007 showed that the change in the global-mean temperature was much more realistic in the ALL ensemble than in the AXA ensemble, which is consistent with the finding from other groups that inclusion of aerosol forcing improves the simulation of twentieth century temperature trends in GCMs.

Figure 3 shows observed and modelled December–February (DJF) rainfall trends for the period 1951–1996. The observed trends in Figure 3(a) clearly show a rainfall increase over the northwest and centre of Australia, which is mainly a summertime phenomenon. The AXA ensemble (Figure 3(b)), forced mainly by increasing greenhouse gases, gives a decrease in summer rainfall in the northwest, in common with many other coupled ocean–atmosphere climate models forced by greenhouse gases (e.g. Whetton *et al.*, 2001, who noted that seven out of eight GCMs actually gave a *decrease* in summer rainfall in the northwest). Inclusion of aerosol forcing in the ALL ensemble (Figure 3(c)) gives an increasing trend of rainfall over much of the continent, and greatly improves the simulated rainfall trends in the northwest. The differences between the rainfall trends in the ALL and AXA ensembles (Figure 3(d)) show that inclusion of aerosols has also improved the simulated rainfall trend in the northeast, even though the model still gives an increasing trend there (whereas the observations show decreasing rainfall over most of the northeast). There are several possible explanations for the discrepancy between modelled and observed rainfall trends, including deficiencies in the model and the effect of natural variability; see R2007, Rotstayn (2007) and Shi *et al.* (2008) for further discussion.

The increase in rainfall in the northwest when aerosol forcing is included correlates with a change in the sign of the trend in meridional surface-temperature gradient in the Indian Ocean sector. As shown in Figure 2, aerosol from Asia essentially remains in the Northern Hemisphere: the impacts on Australian tropical rainfall are not due to Asian aerosol being advected over Australia, but occur because the aerosol cools the Asian continent and surrounding oceans, thus altering the meridional temperature gradient between Asia and Australia. (The cooling effect of aerosols is in contrast with the effect of greenhouse-gas forcing, under which the large Asian continent warms strongly.) The change in temperature gradient alters the monsoonal winds, which bring rainfall to northern Australia. To check whether the simulated rainfall response over Australia was due to Asian aerosol, R2007 performed a sensitivity experiment in which only Asian anthropogenic aerosol sources were allowed to increase with time; this ensemble gave very similar rainfall trends over Australia to those in the ALL ensemble, confirming that the effect was indeed due to Asian aerosol (in the model, at least).

Analysis of the above simulations was extended by Cai *et al.* (2006, 2007) and Cai and Cowan (2007), with a major focus on oceanic processes involved in the response to aerosol forcing. Cai *et al.* (2006) found that, even though most of the aerosol forcing occurs in the Northern Hemisphere, the aerosol-induced change in ocean heat content hardly differed between the two hemispheres during the twentieth century. This was because aerosol-forced cooling of the Northern Hemisphere induced a pan-oceanic northward heat transport, with most of it taking place via a strengthening of the Atlantic meridional overturning circulation. Note that this aerosol-induced strengthening of the Atlantic overturning circulation is in the opposite sense to the greenhouse gas-induced weakening that is often seen in GCMs (Delworth and Dixon, 2006).

Cai *et al.* (2007) compared the output from the CSIRO GCM with a new compilation of historical temperature profiles for the Indian Ocean, the Indian Ocean Thermal Archive (Alory *et al.*, 2007). They found that the strengthening of the Atlantic MOC in the simulation

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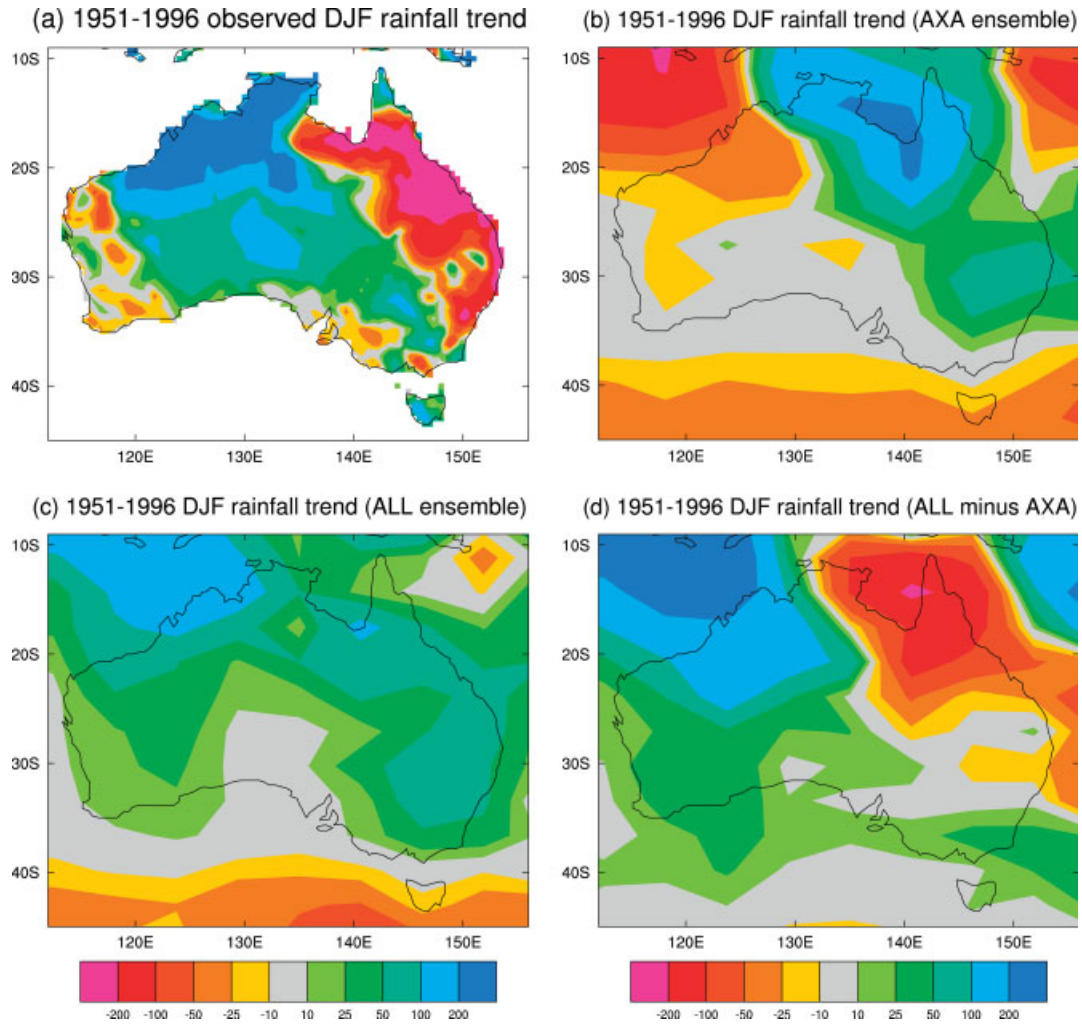

Figure 3. December–February seasonal rainfall trends (mm per century) over the Australian region for the period 1951 to 1996 from observations (Mitchell and Jones, 2005) and from the CSIRO GCM. Modelled trends shown are (b) the AXA ensemble mean, (c) the ALL ensemble mean and (d) the difference between the ALL and AXA ensemble means. Adapted from Rotstayn *et al*. (2007) by permission of the American Geophysical Union. This figure is available in colour online at www.interscience.wiley.com/ijoc

with aerosols increased the warming rate in the southern subtropical Indian Ocean, and by taking heat out of the near-equatorial region, generated a sub-surface cooling there. In both respects, the effects of aerosol forcing improved the agreement of the model with the observed trends from Alory *et al*. (2007). Figure 4(a) shows the observed zonally averaged temperature trends, which are captured much better in the ALL ensemble (Figure 4(b)) than in the AXA ensemble (Figure 4(c)). Anthropogenic aerosol forcing also induced a strengthening and southward shift of the subtropical gyre circulation in the oceans of the Southern Hemisphere. Owing to a positive feedback between the ocean and the atmosphere, this led to a southward shift and strengthening of the westerly winds over the Southern Ocean, and an enhanced positive trend in the southern annular mode (SAM; Cai and Cowan, 2007). (The positive state of the SAM can be defined as an increase in sea-level pressure at 40 °S, and a decrease at 65 °S.)

These results illustrate the power of atmospheric and oceanic teleconnections, which mean that forcing and response can be located far apart (Liu and Alexander, 2007). Effects due to changes in ocean dynamics cannot be captured by atmospheric GCMs coupled to mixed-layer ocean models; this suggests an urgent need for further targeted studies using coupled atmosphere–ocean GCMs.

There are few comparable studies that assess the effects of anthropogenic aerosols in ensembles of twentieth century global climate simulations. In simulations forced only by direct aerosol effects, Delworth and Dixon (2006) noted a marked aerosol-induced strengthening of the Atlantic overturning circulation, similar to what was found in the CSIRO GCM. Although this is encouraging, the above results should be regarded as preliminary. Aside from the simplified treatment of aerosol properties and processes, the low resolution of the CSIRO GCM used in these studies is a cause for concern, since it contributes to a poor simulation of modes of variability such as ENSO (Shi *et al*., 2008). Since the above results suggest that aerosol forcing is crucial for understanding recent trends in the Southern Hemisphere climate, it is

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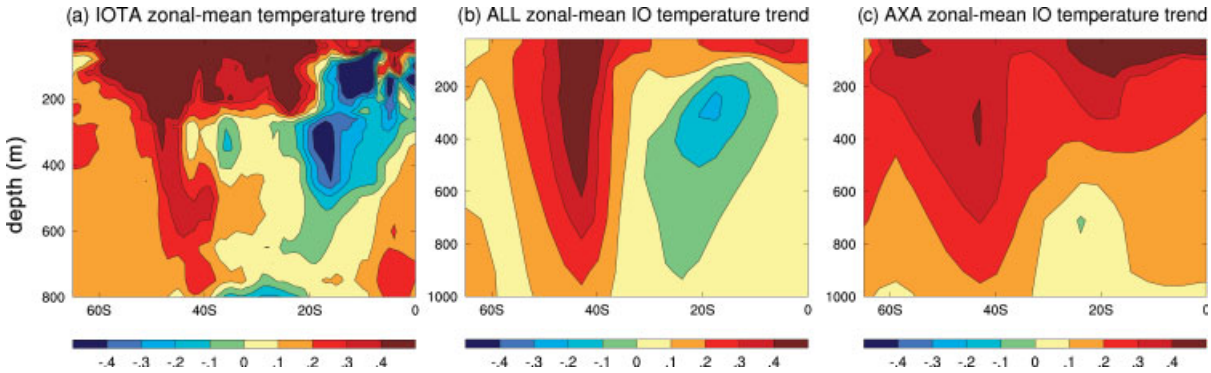

Figure 4. Trends over 1951–2000 in zonal-mean temperature of the upper 800 m of the Indian Ocean from (a) IOTA observations, (b) the ALL ensemble and (c) the AXA ensemble, in units of K per 50 years. Adapted from Cai *et al.* (2007) by permission of the American Geophysical Union. This figure is available in colour online at www.interscience.wiley.com/ijoc

important to determine whether they are reproduced by the high-resolution CSIRO Mk3.5 model or by independent models from other groups.

5. Properties and possible effects of aerosol in the Australian region

There have been few systematic attempts to assess the climatic effects of anthropogenic or natural aerosol perturbations in the Australian region. Existing knowledge from observations and modelling is reviewed in this section. Some existing knowledge is derived from ongoing, coordinated observational programmes. For example, the Australian Bureau of Meteorology and CSIRO operate sun photometers at 16 sites, to characterize the optical properties of Australian continental aerosol (Mitchell and Forgan, 2003). Extension of such measurements from regional to continental scales is being pursued by the retrieval of aerosol properties from satellites (discussed in Section 5.1).

5.1. A top–down view from satellites

Satellite-based remote sensing of aerosol properties is an attractive approach for the extension of surface measurements to the continental and global scales needed to make inferences about effects on climate. Surface measurements, such as those from sun photometers, can be used to validate satellite retrievals (Liu *et al.*, 2004; Kahn *et al.*, 2005), which can in turn be used to validate climate models on broad scales.

The existing work relies mainly on passive instruments. Extracting aerosol information from passive instruments hinges on separation of the solar energy reflected by the aerosol from that reflected by the surface. For single-view sensors, such as MODIS (Moderate Resolution Imaging Spectroradiometer), this requires assumptions about the spectral reflectance properties of the surface that are not well suited to Australian soils and land cover, especially in the arid zone. For this reason, desert dust outbreaks are difficult to identify in the MODIS aerosol product. Multi-angle sensors such as MISR (Multi-angle Imaging Spectroradiometer) and AATSR (Advanced Along Track Scanning Radiometer) are capable of improved aerosol retrieval accuracy over land, since they are better able to differentiate between the surface and atmospheric scattering. MISR can retrieve AOD to typically ± 0.05 (Liu *et al.*, 2004; Kahn *et al.*, 2005). A preliminary study over Australia using MISR and AATSR data from 2004 showed comparable results (Mitchell and Campbell, 2006), although with apparent differences in the slope of the regression line between the satellite retrievals and surface-based measurements relative to the overseas studies. This suggests that the aerosol models assumed in the satellite retrieval algorithms do not provide a sufficiently accurate representation of Australian aerosols. Development of alternative aerosol models suitable for use under Australian conditions is underway.

A promising approach to aerosol remote sensing from satellite uses active instruments such as CALIOP (Cloud Aerosol Lidar with Orthogonal Polarization) on CALIPSO (Cloud Aerosol Lidar Infrared Pathfinder Satellite Observations – Winker *et al.*, 2003). By employing the lidar technique (the optical analogue to radar), these systems have the ability to make direct, rather than inferred, measurements of the altitudes and thicknesses of aerosol layers. They can locate layers by day and by night at vertical resolutions on the order of a few tens of metres and can detect aerosol layers above clouds and, in many cases, below clouds. Because of their relatively small footprints and the ability to discriminate between different layers, the aerosol products from lidar instruments are relatively unaffected by the cloud clearing and cloud contamination problems that affect some passive techniques.

The primary outputs from CALIOP are two-dimensional, height *versus* along-track distance 'curtain files' of attenuated backscatter. As an example, the 1064-nm attenuated backscatter measured over Australia is shown in Figure 5. This figure shows aerosols lofted to over 6 km near the centre of Australia, where the mixed layer is capped by convective clouds (white in the figure) that attenuate the lidar signal. Ice clouds between 10 and 14 km appear less attenuating. An intense aerosol event (brighter shading around 13Â°S) located over northern

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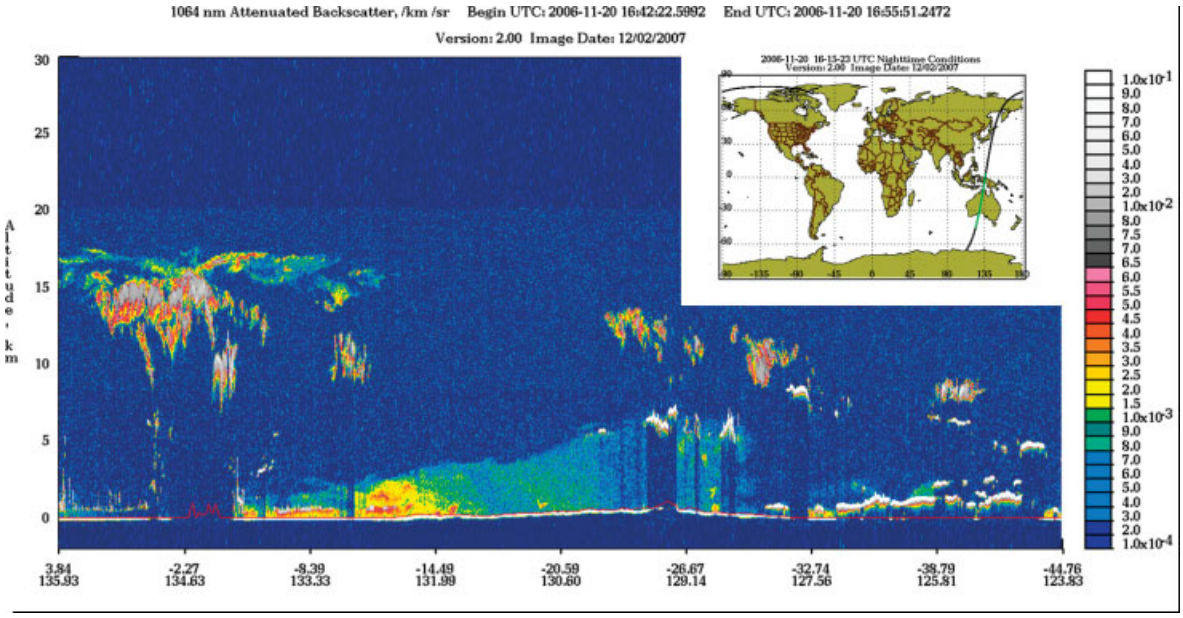

Figure 5. 1064-nm attenuated backscatter measured by the CALIPSO lidar on 20th November 2006. The units of the attenuated backscatter shown in the scale bar are (kilometre steradian)-1. The lighter section of the ground-track line in the insert shows the extent of the CALIPSO data depicted in the main figure. The figures were obtained from the NASA Langley CALIPSO website: <http://wwwcalipso.larc.nasa.gov/products/lidar/>. This figure is available in colour online at www.interscience.wiley.com/ijoc

Australia (at ~13 °S) is associated with the smoke from many fires burning in the area. CALIOP also measures the polarization components of the backscattered signal to assist in the classification of aerosol layers detected in the troposphere as either biomass burning smoke, desert dust, clean marine, clean continental (background), polluted continental or polluted dust (Omar *et al*., 2004; Liu *et al*., 2008).

CALIOP data are being used to create climatologies of the global, three-dimensional distribution of aerosols and clouds, aerosol optical properties and aerosol types. CALIOP Level 2 data products include vertical profiles and layer values of aerosol and cloud extinction coefficient and optical depths. This information is required for the calculation of the direct radiative effects of aerosols and their effects on clouds.

5.2. Aerosol from Australian biomass burning

Every year, biomass burning releases large quantities of aerosol that are significant at both regional and global scales. In terms of global total carbon emissions, it is estimated that biomass-burning emissions are approximately one-third to one-half of those from fossil-fuel combustion, and that savannah fires are responsible for about 50% of the total emissions from biomass burning (Williams *et al*., 2007). According to a database used by van der Werf *et al*. (2006), the global annual biomass burning emissions of total particulate matter and $PM_{2.5}$ (particulate matter with an aerodynamic diameter of 2.5 μm or less), averaged over the years 1997–2006, were 50 Tg and 37 Tg respectively. Of these, the Australian contributions were 5.5 and 4.6%, respectively. Most of the Australian emissions result from wild fires and prescribed fires in the savannah regions of tropical Australia (Kasischke and Penner, 2004).

Several authors have considered the direct radiative effects of the carbonaceous aerosol produced by biomass burning during the dry season (April–November) in northern Australia. Sun-photometer measurements at the most northern tropical stations show a clear annual cycle in mid-visible AOD, with a peak monthly mean AOD typically around 0.5 during September–October (Radhi *et al*., 2006). O'Brien and Mitchell (2003) combined satellite retrievals with surface-based sun-photometer observations to calculate the aerosol-radiative heating rates at Jabiru in Kakadu National Park, and found that the radiative heating was substantial, even in comparison with the heating produced by water vapour.

Spatio-temporally resolved fields of aerosol emissions due to biomass burning are essential for modelling and assessing emissions' impact on climate and weather, and several researchers have developed such fields at global scale using a combination of satellite and ground-based measurements and semi-empirical methods (e.g. Ito and Penner, 2004; van der Werf *et al*., 2006). Recently, biomass burning emissions in northern Australia were considered in a two-part study by Meyer *et al*. (2008) and Luhar *et al*. (2008). Meyer *et al*. constructed a high-resolution inventory of aerosol emissions for the Top End (the monsoonal top half of the Northern Territory) for the 2004 dry season, and validated the emissions using a three-dimensional meteorological and transport model (The Air Pollution Model (TAPM); Hurley *et al*., 2005). They derived the emissions using information on fuel-load distribution, satellite-based measurements of fire scars and hot spots, and a version of the McArthur fire danger meter (an empirical measure of expected fire

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behaviour for a standard fuel type). The total emission of $PM_{2.5}$ from the Top End for the 2004 season computed by Meyer *et al.* was 0.67 Tg.

Luhar *et al.* used the above aerosol emission rates in TAPM, assuming no secondary aerosol generation, and compared the modelled aerosol concentrations and AODs with satellite and ground-based measurements, obtaining generally satisfactory results. Their estimated top-of-atmosphere radiative forcing due to aerosol has a seasonal mean of -1.8 W m^{-2}, with a region of strong enhancement over the western portion of the Top End (Figure 6). Given that this radiative effect occurs during the dry season, it is unclear whether these emissions affect regional climate or rainfall, but further study is warranted. In particular, in view of the deficiencies noted above with regard to the biomass-burning emissions used by the CSIRO GCM over northern Australia, the possibility of extending the high-resolution emissions methodology of Meyer *et al.* (2008) to a wider domain is attractive.

Little attention has been paid to smoke emissions from bushfires in Australian temperate forests. However, a recent study indicated that such episodic events have very large radiative effects. Mitchell *et al.* (2006) used sun-photometer measurements to determine the radiative impact of the bushfire smoke (aerosol) generated by the Canberra firestorm of January 2003. They estimated mean radiative forcings of -50 W m^{-2} at the top of the atmosphere and -172 W m^{-2} at the surface during the week following the firestorm. The smoke layer stabilized the lower troposphere through a combination of reduced surface heating and a positive gradient in heating rate through most of the layer. The enhanced stability suppressed surface temperatures and prevented the north-westerly winds from penetrating to the surface during the week following the firestorm. This reduced the risk of renewed fire danger, and suggests an important role for inclusion of aerosol effects in weather prediction models.

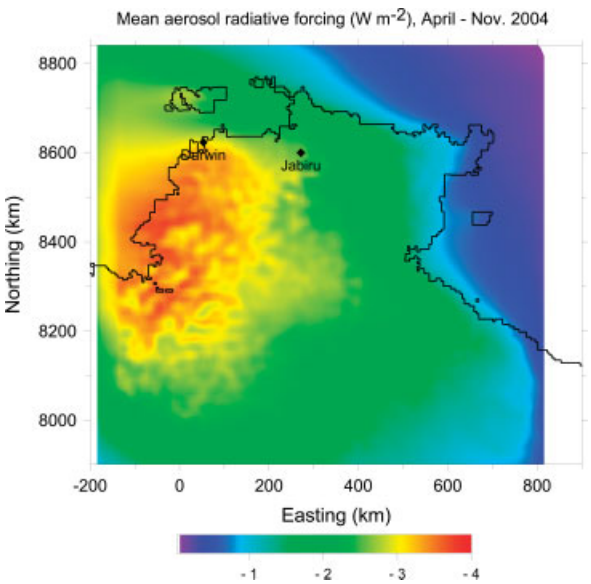

Figure 6. Distribution of aerosol-radiative forcing (W m^{-2}) over the Top End, averaged over the period April–November 2004 (from Luhar *et al.*, 2008). This figure is available in colour online at www.interscience.wiley.com/ijoc

5.3. Aerosol from Indonesian biomass burning

The aerosol source from Indonesian biomass burning is larger than any in Australia, and it displays large inter-annual variability, due to the correlation with ENSO. During strong El Niño events, Indonesia can represent a large fraction of the global source of aerosol from biomass burning. For example, Davison *et al.* (2004) estimated the total particulate matter emission from Indonesian forest fires for August–November 1997 at 41 Tg (with a large uncertainty range of 17–76 Tg). This is substantial compared to estimates for the annual average global emission from all sources of biomass burning of 50 Tg as given above, and 60 Tg from Penner *et al.* (2001). Smaller values of 8.3 and 21 Tg for the aerosol emissions from the Indonesian fires were estimated in two earlier studies; see Davison *et al.* (2004) for discussion. Indonesian aerosol contains a substantial amount of sulfate from the combustion of subterranean peat, which is thought to make the aerosol relatively hygroscopic compared to that from other biomass-burning sources (Gras *et al.*, 1999). This is likely to enhance both the direct and indirect effects of Indonesian biomass-burning aerosol.

Figure 7 shows the change in incident short-wave flux at the surface due to biomass-burning aerosol in September 1997, as calculated with the Met Office's atmospheric model by Davison *et al.* (2004), using their best estimate of the aerosol emissions. They simulated large reductions of up to 200 W m^{-2} over Indonesia, with smaller reductions over the Indian Ocean and northern Australia. Their results are broadly consistent with another modelling study by Duncan *et al.* (2003), who obtained reductions of up to 178 W m^{-2} over Indonesia. These studies considered only the direct effects of aerosol on solar radiation, and did not attempt to calculate indirect effects,

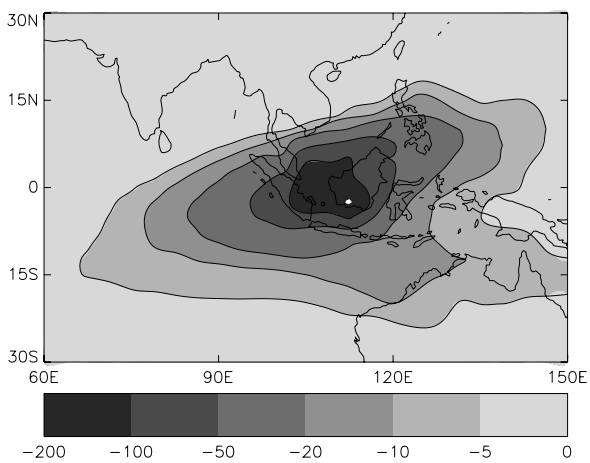

Figure 7. Simulated change in incident short-wave flux at the surface (W m^{-2}) for September 1997 due to biomass-burning aerosol. Reproduced from Davison *et al.* (2004) by permission of the American Geophysical Union.

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or the climatic response to the forcing. Recently, Graf *et al*. (2007) simulated the effects of the Indonesian fires on convective clouds and precipitation during September 1997 using a regional model, and found that convective precipitation was reduced at all places with high aerosol loading. These studies are suggestive of large climatic effects due to Indonesian biomass burning. In particular, the increase of burning in El Niño years such as 1997 and 2006 could give rise to positive feedbacks on the natural ENSO cycle.

5.4. Dust aerosol from wind erosion of soils

Dust is both strongly influenced by climate and itself influences climate. There is a growing body of evidence from overseas studies that dust has substantial impacts on short- and long-wave radiation. For example, Haywood *et al*. (2003) estimated a maximum decrease in short-wave radiative at the surface of 209 W m^{-2} during a dust event over the North Atlantic Ocean in September 2000. As a partially absorbing aerosol, dust tends to increase atmospheric stability and reduce convection (Satheesh and Moorthy, 2005). The IPCC Fourth Assessment estimates the global-mean radiative forcing of anthropogenic dust as -0.1 [± 0.2] W m^{-2} (Forster *et al*., 2007). This relatively low value occurs because the anthropogenic component is estimated at between 0 and 20%, and due to cancellation between short- and long-wave radiative effects. Dust may also be important as an active player in climate variability (e.g. in modulation of tropical cyclone activity) and as a biogeochemical link between land, air and sea (Ridgwell, 2002), in addition to the anthropogenic radiative forcing as defined by the IPCC.

Australia is the dominant source of dust in the Southern Hemisphere (Tanaka and Chiba, 2006; Mackie *et al*., 2007). Figure 8, from the global model of Tanaka and Chiba, suggests that Australia contributes more than 70% of the atmospheric dust loading over most of the Southern Hemisphere.

Compared to the major Northern Hemisphere dust source regions, Australian dust emissions are low and episodic. Despite this, Australia's Southern Hemisphere location and the sensitivity of its responses (in both time and space) to changing climate increases the continent's importance as a source of dust and possibly also increases the importance of Australian dust as a climate driver. However, there have been few direct measurements of the radiative effects of Australian dust.

A 45-year meteorological record of dust activity (1960–2005) for the continent is provided by the Dust-Storm Index (DSI) of McTainsh *et al*. (2007). This record highlights the highly episodic nature of Australian dust activity, which is strongly controlled by rainfall. The three main periods of high activity; 1960–1972, 1994 and 2002–2003 are all associated with droughts. This continent-wide record is however smoothed by the tendency for the east and west of the continent to have quite different temporal patterns of activity; a feature which global dust models have difficulty describing (Tegen *et al*., 2002; Tanaka and Chiba, 2006). Not only does this temporal and spatial complexity produce highly variable atmospheric dust loads, which will have radiation implications, the complex patterns of dust source areas yielding dust colours ranging from red to white could also produce 'warmer' and 'colder' dusts. Also, variations in organic matter (Boon *et al*., 1998) and aggregation (Leys *et al*., 2005) will affect dust radiative properties. As noted by the IPCC (Forster *et al*., 2007), the radiative effects of dusts from source regions with variable mineralogies is generally not described by global models.

Saharan dust-induced surface cooling and lower atmospheric heating has been shown to increase atmospheric stability and reduce convection in the tropical Atlantic

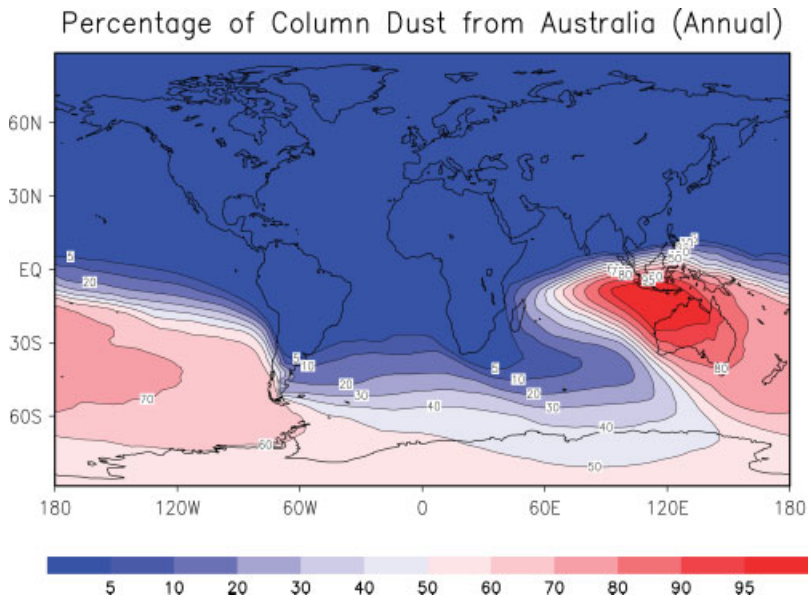

Figure 8. Contribution (in percent) of the annually averaged column-integrated dust mass from the Australian source region. Adapted from Tanaka and Chiba (2006) by permission of Elsevier Limited. This figure is available in colour online at www.interscience.wiley.com/ijoc

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Ocean and this may explain the strong inverse relationship between inter-annual variations of tropical cyclone activity and Saharan dust outbreaks (Evan *et al.*, 2006; Lau and Kim, 2007a,b). The distant ends of the two major dust paths exiting the Australian continent – to the west (over the Northwest Shelf) and east (over the Coral Sea) – are also the focii of tropical cyclone activity. There is therefore potential for Australian dust outbreaks to affect cyclone activity in the Australian region.

A larger number of dust plumes exiting the east of the continent track to the south into the Southern Ocean (McGowan *et al.*, 2000). The deposition of iron-rich dusts from these plumes could play a critical role in fertilization of the Southern Ocean (Boyd *et al.*, 2004; see Section 5.5).

5.5. Iron fertilization of the Southern Ocean by dust

Dust deposition over the oceans provides an important biogeochemical link between terrestrial and marine ecosystems that is likely to be influenced by climate change (Ridgwell, 2002). In 'high nutrient–low chlorophyll' ocean regions, such as the Southern Ocean, iron and other micronutrients are seen as limiting primary production, and thus affecting atmospheric CO_2 concentrations.

Australian mineral dust is high in iron content and can be transported over the Subantarctic Southern Ocean, particularly during the austral spring and summer when seasonal dust-storm frequency in southern Australia is highest (Boyd *et al.*, 2004). Figure 9 shows simulated dust deposition to the surface from the GOCART model (Ginoux *et al.*, 2001) for November, 2002, during the very active dust-storm season of 2002–2003. Gabric *et al.* (2002) reported a coupling between satellite-derived AOD (affected by dust aerosol) and chlorophyll concentration (a proxy for phytoplankton biomass) in the upper ocean, providing evidence that episodic delivery of wind-blown iron was stimulating phytoplankton growth in this region. This was corroborated by Cassar *et al.* (2007), who showed that net primary production over large areas of the Subantarctic Southern Ocean is proportional to dust-derived iron deposition.

There is evidence that dust deposition to the Southern Ocean affects atmospheric CO_2 concentration on both glacial and inter-annual timescales. It was hypothesized by Martin (1990) that dust levels during the last glacial period increased the supply of iron to the oceans, with a consequent draw-down of atmospheric CO_2. This is supported by Antarctic ice cores (Petit *et al.*, 1999; Edwards *et al.*, 2006) and by a number of modelling studies (e.g. Ridgwell, 2002; Mahowald *et al.*, 2006b). In this view, dust was an active player in glacial-interglacial climate transitions. An ice core from East Antarctica indicates that deposition of iron was unusually large in 1927–1928, which coincided with a severe drought and increased export of dust from Australia (Edwards *et al.*, 2006). More frequent or severe Australian droughts under climate change could thus contribute to a decrease in atmospheric CO_2 levels. Conversely, reafforestation could

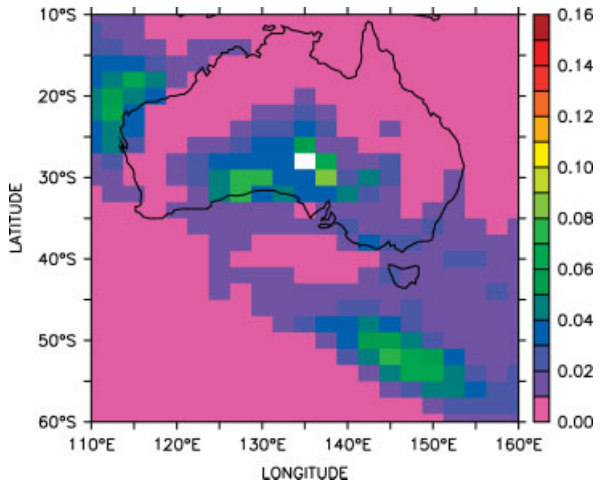

Figure 9. Monthly mean dust deposition for November 2002 (in $\mu g\ m^2\ s^{-1}$) from the GOCART dust transport model. This figure is available in colour online at www.interscience.wiley.com/ijoc

weaken the oceanic carbon sink and potentially offset some of the carbon sequestered on land. The dust–CO_2 connection is an excellent example of the feedbacks that can occur in the interconnected Earth system (Ridgwell, 2002).

5.6. Natural aerosol over the Southern Ocean

Australia's isolated location gives natural aerosol a relatively important role over much of the continent. Southern Ocean boundary-layer processes scrub pollutants from air reaching the southern coasts, and marine aerosol can traverse significant distances inland. In mass terms, the dominant process is sea-salt generation, which is wind-speed dependent and highly non-linear. Mass emission is most useful in considering effects such as nutrient transport into continental areas, for example, salt transport: in Western Australia, chloride deposition is around an order of magnitude greater in the south-west than in the north, and marine influence is evident over 200-km inland (Hingston and Gailitis, 1976; Keywood *et al.*, 1997). At cloud level, sea-salt particles, which can be taken as a proxy for all marine-generated particles, penetrate significantly further. For example, King and Maher (1976) reported a reduction of only a factor of 5 in concentration over 1100 km from the coast in central Queensland. This is also consistent with earlier findings by Twomey (1955) of a lack of significant decrease in particle concentration (at altitude) over south-eastern Australia in the absence of precipitation.

Measurement of total particle number concentration and concentration of particles active as CCN over three decades at Cape Grim, Tasmania, together with particle composition (methanesulfonate (MSA) and non–sea-salt sulfate) and precursor (DMS) concentrations have revealed very systematic underlying seasonal patterns showing summer maxima and winter minima. The predominant source of these nuclei is believed to be the oxidation of DMS from marine phytoplankton, although a

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small fraction of CCN (possibly ~50 cm^{-3}) appear unrelated to this process and can be attributed, at least in part, to sea-salt (Ayers and Gras, 1991).

Long-term particle measurements at Cape Grim have revealed strong inter-annual variations in total particle number concentrations and CCN, with total particle number concentration increasing by $2.4 \pm 0.3\%$ per year but for CCN decreasing by $1.2 \pm 1\%$ per year. These trends suggest a shift in particle production/loss mechanisms between 1997 and 2003, resulting in more small particles that do not significantly contribute to CCN. The underlying mechanism for these trends is unclear and is the subject of continuing research.

The role of natural aerosol over the Southern Ocean in climate-change feedbacks is largely unquantified, but preliminary evidence suggests that it could be important. The possibility that DMS-derived aerosol could regulate climate by increasing CCN concentrations in response to changes in temperature or solar energy was proposed more than 20 years ago by Charlson *et al.* (1987), the so-called CLAW hypothesis. Latham and Smith (1990) suggested the possibility of a similar negative feedback due to increased levels of sea-salt in response to an increase in wind speed under enhanced greenhouse conditions. On the basis of measurements of stratocumulus clouds over the Southern Ocean, Boers *et al.* (1998) found markedly higher cloud-droplet number concentrations and cloud albedos in summer than in winter, and attributed the difference to CCN derived from DMS. This suggests that DMS-climate feedbacks are likely to be mediated by indirect aerosol effects as well as direct radiative effects.

Some recent studies have attempted to quantify aspects of these feedback loops in global models. For example, Gabric *et al.* (2004) used a marine food-web model, the CSIRO Mk2 atmosphere-ocean GCM, and an empirical DMS algorithm to predict the DMS seawater concentration and the DMS sea-to-air flux under contemporary and enhanced greenhouse conditions. At the time of equivalent-CO_2 tripling, the largest increase in DMS flux (106%) was simulated in the Southern Hemisphere between 50 and 60 °S, mainly due to GCM-simulated decreases in the ocean mixed-layer depth. However, using another GCM, Kloster *et al.* (2007) found that the largest *decrease* in the DMS seawater concentration under climate change was in the Southern Ocean (−40%), due to an *increase* in the summer mixed-layer depth. This shows that the sign of future DMS changes in the Southern Ocean is currently unknown. Climate-change simulations by Mahowald *et al.* (2006a) showed marked increases in sea-salt aerosol loading south of 50 °S in response to a doubling of CO_2, due to increased wind speed at high latitudes in the Southern Hemisphere. This increase in wind speed (combined with a decrease at mid-latitudes) is one of the most consistent responses of GCMs to an increase in greenhouse gases. The poleward shift of the Southern Hemisphere storm tracks appears to be at least partly related to changes in meridional temperature gradient: there is enhanced warming in the tropical and subtropical upper troposphere, due to a smaller moist adiabatic lapse rate in a warmer climate, and relatively little warming near the surface poleward of about 40 °S, due to the large thermal inertia of the Southern Ocean (Yin, 2005). The poleward shift of the storm tracks is also qualitatively consistent with the observed trend of the SAM towards its positive state in recent decades. Thus, feedbacks involving natural aerosol, the SAM, and changes in temperature, solar energy and ocean mixed-layer depth may have a significant role in modulating climate change in the Southern Ocean and over southern Australia. However, the physical and chemical processes involved are still poorly understood, and it is unclear whether DMS-derived sulfate is indeed the dominant source of CCN in the marine boundary layer (e.g. Cainey *et al.*, 2007).

5.7. Biogenic VOC emissions and aerosol

Volatile organic compounds (VOCs) emitted from vegetation can react in the atmosphere and condense to form new particles or supplement existing aerosol. It was argued by Crutzen *et al.* (1999) and subsequently substantially documented by Kesselmeier *et al.* (2002) that a significant fraction of the carbon fixed as net primary productivity of an ecosystem is lost as VOCs from the plants to the atmosphere. The rates of VOC loss are both species- and climate-dependent. Very little work has been done on VOC emissions from Australian vegetation, although the smell of terpenes and eucalyptus oil vapour in forested areas on a hot day is well known. Once these VOCs are in the atmosphere, selected species among them form SOA. It has been argued that these are the dominant aerosol in the free atmosphere (Goldstein and Galbally, 2007). Furthermore, recent research shows that, while fresh SOA is not hydrophilic (Varutbangkul *et al.*, 2006), aged SOA has light scattering and cloud-droplet forming properties similar to inorganic salts (Dinar *et al.*, 2006), making this SOA climatically active not just through the direct effect but also through the indirect effects. This SOA is evident in Australia in summer as the blue haze that is exemplified in the Blue Mountains. Scaling the global estimates of SOA production of Goldstein and Galbally (2007) to Australia suggests that continental SOA production may be in the range 3–10 Tg/year, which would make this the major and least studied aerosol source in Australia.

5.8. Non-episodic continental aerosol

Little is known about natural production aerosols over Australia (i.e. aerosol that is not associated with regional-scale fires and dust). Over most of the continent, particle concentrations in the boundary layer are very low (generally less than 1000 cm^{-3}) and in clean areas they are typically 250–500 cm^{-3}, indicating very few active surface sources (Bigg and Turvey, 1978). However, Bigg and Turvey did not specifically examine forested areas and there is no subsequent information that indicates how these concentrations may have changed over the past three decades.

A number of short-term measurement campaigns have assessed non-episodic aerosol concentrations at different

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locations across Australia, all suggesting extremely low concentrations of aerosol mass (typically in the range 0.1–5 μg m^{-3}) and very low scattering coefficients (Gras *et al*., 1999; Carr *et al*., 2005). Information on the composition and size of these natural aerosol is limited; Ikegami *et al*. (2004) and Okada *et al*. (2008) show that over 90% of free-tropospheric particles between 0.2 and 1–2 μm diameter were sulfate or sulfuric acid, with small contributions of mineral-dust or sea-salt particles. Over central Australia, roughly 30–50% of sulfur-rich particles were also potassium-rich (hence smoke-related) or mineral-rich. This points to the potential for a significant fraction of free-tropospheric particles over the continent to contain some surface-derived primary components.

During August to November, the situation is different because much of the continent is affected by biomass-burning smoke transported from northern Australia and elsewhere in the Southern Hemisphere. Balloon soundings from Mildura between 1972 and 1980 by Gras and Laby (1978, 1979a,b) showed dramatic increases in concentrations of fine particles from the surface to about 10 km. Rosen *et al*. (2000) confirmed the large horizontal scale of laminar transport during spring at Mildura and show the likely origin of the enhancements as advected smoke from either Africa or South America. The importance of long-range transport of biomass-burning emissions from southern Africa is further shown from observations by Pak *et al*. (2003) over south-eastern Australia. Aerosol properties were also observed in the mid-troposphere over central Australia in October 1994 (Okada *et al*., 2005). For particles 0.2–0.7 μm in diameter, 88% were internally mixed soot, thought to be aged biomass-burning particles transported from northern Australia.

5.9. Possible modulation of rainfall by Australian anthropogenic aerosol

Cloud-physics theory has long suggested the possibility of rainfall suppression in warm clouds affected by pollution aerosols that act as CCN, since this may lead to smaller cloud droplets and reduced coalescence of droplets (e.g. Squires, 1958). Recently, it has been recognized that under moist, unstable conditions, anthropogenic aerosols may actually invigorate deep clouds and lead an overall increase in rainfall (e.g. Rosenfeld, 2006; Bell *et al*., 2008). Thus, the effect of aerosol pollution on rainfall is likely to depend on the precipitating environment, as well as the type of aerosol.

A number of authors have considered the possibility of rainfall suppression in Australian clouds affected by anthropogenic pollution. Warner and Twomey (1967) analysed data taken upwind and downwind of massive sugar-cane fires in Queensland, and showed that the cloud-droplet concentration was clearly increased in clouds affected by the smoke aerosol from the fires. However, subsequent comprehensive analysis of over 60 years of rainfall records failed to reveal an association between decreases in rainfall downwind of the sugar cane fires and expanding cane production (Warner, 1968, 1971).

More recently, Rosenfeld (2000) presented satellite retrievals of cloud-droplet effective radius and precipitation formation for two days over south-eastern Australia, and argued that these showed suppression of precipitation in clouds affected by aerosol pollution. In particular, he suggested that observed decreases in precipitation in the Snowy Mountains during the twentieth century could be due to aerosol pollution. However, Nicholls (2000) showed that an apparent strong downward trend in the district average rainfall for the Snowy Mountains since 1913 was an artificial result of changes in the mix of stations used to produce the district average. Further, Nicholls (2005) showed that, although the snow season in the Australian Alps was shortening, with less snow remaining early in spring, this was due to warming rather than any substantial decline in precipitation. IOCI (2002) considered local aerosol pollution in its detailed study of the rainfall decline in southwest Western Australia, and concluded that it was unlikely to be a major contributing factor. Some other aspects of Rosenfeld's analysis have been strongly debated (Ayers, 2005; Rosenfeld *et al*., 2006), and the issue has also generated controversy in Israel (Givati and Rosenfeld, 2004, 2005; Alpert *et al*., 2008).

Recently, Bigg (2008) analysed rainfall records in the area around Brisbane and the Gold Coast, and found that the spatial distribution of rainfall trends was consistent with the likely distribution of aerosol pollution from human activity. He argued that aerosol observations in the area are urgently required, in order to validate models and apportion the causes of rainfall changes.

In summary, validation of rainfall change due to aerosol pollution remains a contentious issue.

6. Summary and key research directions

This review was motivated by strong evidence that aerosol exerts substantial effects on climate. This evidence is especially compelling in the Northern Hemisphere, where aerosol concentrations are larger than in the Southern Hemisphere. However, new exploratory climate modelling with a low-resolution version of the CSIRO GCM (Rotstayn *et al*., 2007) suggests that aerosol effects are of comparable importance to greenhouse gases as a driver of recent climate trends in the Southern Hemisphere, including Australia. The large simulated impact of anthropogenic aerosol is principally due to an inter-hemispheric contrast in aerosol forcing, which induces changes in atmospheric and ocean circulation. These changes are hypothesized to have affected Australian tropical rainfall, temperature trends in the Indian Ocean and the behaviour of the SAM. There are currently few studies with other coupled ocean-atmosphere GCMs that can be used for comparison.

Our review also considered the possible climatic effects of natural and anthropogenic aerosol located in the Australian region. Observations and modelling suggest that various regional aerosol sources may be important,

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but systematic studies of their role in climate change and climate variability are currently lacking. Examples include the large modelled reductions in short-wave flux at the surface due to biomass-burning events in Australia and Indonesia, and feedbacks involving natural aerosol processes over the Southern Ocean.

Understanding and having some predictive capacity of climate variability and change is a stated national priority goal associated with an environmentally sustainable Australia. The above results from the CSIRO GCM demonstrate that a better understanding of aerosol effects is essential in order to reconcile modelled and observed climate variability and climate change. Although greenhouse-gas forcing is expected to eventually become the dominant driver of climate change during the twenty-first century, it will be difficult to predict regional climate change without improved knowledge of the climatic effects of aerosols.

Models are only as good as the information that goes into them. This information includes aerosol emission inventories and parameterization of aerosol physical and chemical processes. Strong linkages are required to connect field and laboratory measurements to the simplified treatments used in climate models; detailed process models and satellite retrievals can provide these links, and can be used to validate climate models. A comprehensive strategy of this type was outlined by Ghan and Schwartz (2007).

Our recommendations for further research in Australia are as follows:

1. The existing CSIRO Mk3 GCM, with only minor modifications, is a valuable tool for exploring some of the hypotheses raised by the low-resolution simulations described above, and should be exploited more fully.

In particular, do high-resolution (spectral T63) simulations give similar results to the low-resolution version? The T63 version, with horizontal resolution of roughly 1.9° by 1.9°, is better able to resolve oceanic circulation, and has a much better representation of ENSO-related variability than the low-resolution version (roughly 5.6° in longitude by 3.2° in latitude). With this in mind, the aerosol treatment has been extended to the high-resolution version of the GCM, which is currently being tested and tuned. Other related hypotheses can also be explored with sensitivity tests using the existing model, even at low resolution. For example, do wind-related natural aerosol feedbacks in the Southern Ocean potentially exert a positive feedback on the SAM? This question also leads naturally to the topic of aerosol–cloud interactions, since it is principally through cloud-albedo changes that DMS-related feedbacks are hypothesized to operate (Section 5.6). The simplified treatments in the GCM allow only a limited exploration of the possible feedbacks, but demonstration of such a sensitivity would provide a focus for future work.

2. Development of better aerosol treatments for Australian climate modelling will require measurement programmes that are carefully integrated with process-oriented models and GCMs.

Future climate modelling in Australia will be carried out using the Australian Community Climate Earth-System Simulator (ACCESS), a joint initiative of the Bureau of Meteorology and CSIRO in cooperation with the university community (http://www.accessimulator.org.au). The atmospheric component of ACCESS is based on the Met Office's Unified Model, which employs similar simplifications in its aerosol treatment to those in the CSIRO GCM. However, a new aerosol treatment, which predicts both mass and number concentration of various aerosol modes, is currently under development in the United Kingdom (Spracklen *et al.*, 2007). It is expected that future development of this model will benefit from observational and process-oriented aerosol work carried out around the world. It is thus recommended that research in Australia should focus on a limited number of topics that are likely to be of regional climatic significance, and where the capacity exists in Australia to carry out internationally competitive research with benefits for ACCESS and other modelling efforts. The following are identified as priority areas. If preliminary studies suggest that Australian sources are climatically significant, then a substantial research effort would be justified.

- Natural aerosol over the Southern Ocean: This is a priority because of Australia's proximity to this uniquely clean environment, and positive feedbacks that may occur between natural aerosol and atmosphere-ocean dynamics under climate change. Important aerosol types include natural sulfate and organics, sea-salt and wind-blown dust (for its role in iron fertilization of the Southern Ocean). Marine boundary-layer clouds are highly susceptible to perturbed aerosol concentrations, so indirect aerosol effects are likely to be important.
- Tropical biomass burning in Australia and Indonesia: This is potentially important due to substantial radiative effects, especially in El Niño years, and the poor treatment of emissions from this region in current inventories. There is scope to extend the emissions methodology of Meyer *et al.* (2008) to a wider area, including Indonesia, and there is also scope to undertake observational work in Indonesia, probably in collaboration with overseas groups. Possible interactions with convection suggest synergies with other Australian research into moist tropical processes.
- Biogenic secondary organic aerosol from VOCs: It is now recognized that this natural source of secondary organic aerosol is seriously underestimated in standard models, and the underlying chemical mechanisms have recently become an active area of research. It is currently unclear whether the Australian source is climatically significant.
- Dust, for its possible role in climatic variability via direct radiative effects: Saharan dust has recently been identified as a key factor that modulates inter-annual variations in Atlantic tropical cyclones,

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but it is currently unclear whether Australian dust loadings are high enough to exert such effects.

- Modulation of rainfall by anthropogenic aerosol: This issue has been contentious, both in Australia and elsewhere. In view of the large socioeconomic impacts of drought in Australia, further studies seem justified to clarify whether the issue is important in climatic terms.

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