Global connections between aeolian dust, climate and ocean biogeochemistry at the present day and at the last glacial maximum

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Abstract

Palaeo-dust records in sediments and ice cores show that wind-borne mineral aerosol ('dust') is strongly linked with climate state. During glacial climate stages, for example, the world was much dustier, with dust fluxes two to five times greater than in interglacial stages. However, the influence of dust on climate remains a poorly quantified and actively changing element of the Earth's climate system. Dust can influence climate directly, by the scattering and absorption of solar and terrestrial radiation, and indirectly, by modifying cloud properties. Dust transported to the oceans can also affect climate via ocean fertilization in those regions of the world's oceans where macronutrients like nitrate are abundant but primary production and nitrogen fixation are limited by iron scarcity. Dust containing iron, as fine-grained iron oxides/oxyhydroxides and/or within clay minerals, and other essential micronutrients (e.g. silica) may modulate the uptake of carbon in marine ecosystems and, in turn, the atmospheric concentration of CO_2. Here, in order to critically examine past fluxes and possible climate impacts of dust in general and iron-bearing dust in particular, we consider present day sources and properties of dust, synthesise available records of dust deposition at the last glacial maximum (LGM); evaluate the evidence for changes in ocean palaeo-productivity associated with, and possibly caused by, changes in aeolian flux to the oceans at the LGM; and consider the radiative forcing effects of increased LGM dust loadings.

Keywords: aerosols, dust, climate change, palaeoclimatology, radiative forcing, iron fertilisation.

1. *Introduction*

Palaeo-dust records show that wind-borne mineral aerosol (here referred to as 'dust') is strongly linked with climate state. Studies of dust in sediments and ice cores show that dust concentrations and fluxes have changed greatly in association with changes in climate, for example, in the transitions from glacial to interglacial regimes. During glacial climate stages, the world was much dustier, with dust fluxes two to five times greater than in interglacial stages (e.g. Kohfeld and Harrison, 2001). However, the influence of dust on climate remains a poorly quantified and actively changing element of the Earth's climate system. We know from present-day studies that dust can influence climate directly, by changing the radiative properties of the atmosphere through the scattering and absorption of solar and terrestrial radiation, and indirectly, by acting as ice nuclei (Sassen et al., 2003) and modifying cloud properties which, in turn, can impact both the radiative balance of the Earth and the hydrological cycle (Arimoto, 2001). Dust transported to the oceans can also affect climate indirectly, by supplying elements such as Fe, an essential micronutrient in enzymes essential to photosynthesis (Martin et al., 1991); phytoplankton in about 40% of the world ocean are Fe-limited (Moore et al., 2002). Thus, dust delivery to the ocean can modulate the uptake of carbon in marine ecosystems and, in turn, the atmospheric concentration of CO_2.

Dust not only can affect climate, but the generation and transport of dust is itself extremely sensitive to climate. At the present day, the most obvious link is with aridity – the globally dominant sources of dust are all located in arid or semi-arid regions. Global model estimates of present-day dust mobilization rates are poorly constrained, reflecting the scarcity of temporal and spatial data coverage, and range between ~ 1 and 3.5 Pg yr^{-1} (e.g. Tanaka and Chiba, 2006; Zender et al., 2003; Engelstaedter et al., 2006); optimized multi-model estimates yield a range of 1.5 to 2.6 Pg yr-1(Cakmur et al., 2006). Estimates of deposition to the oceans range from about 0.3 to 2 Pg yr^{-1} (Zender et al., 2004; Mahowald et al., 2005). Human activities can have an impact on dust mobilization but here too estimates are poorly constrained and range from 0 to 50 % of global dust emissions (e.g. Tegen et al., 2004; Mahowald et al., 2004; Yoshioka et al., 2005).

As recorded in sediments and ice cores, there have been large and systematic variations in dust loading in the past. The large changes in dust emissions and transport seen from the palaeo-dust record may reflect a variety of processes: changes in sources and source conditions; changes in vegetative cover; sub-aerial erosion of emergent continental shelves; deflation from periglacial deposits; variations in wind speed and gustiness; changed wind patterns linking sources to deposition areas; changes in deposition along the dust transport path. In order to interpret the palaeo-record and to anticipate future changes in climate, we need to have a better understanding of the factors that affect dust mobilization and its subsequent climatic impacts.

The complex linkages between climate and the dust generation-transport process on glacial-interglacial time scales is illustrated in figure 1. There are many processes that can affect the dust cycle either directly or indirectly under any specific climate scenario. The relative importance of any specific process can change dramatically when shifting from one climate state to another – e.g., during glacial stages, low sea levels expose coastal sediments and glacial action can produce unvegetated, unconsolidated deposits all of which can become major sources of dust. A major driver in the Earth's climate system is atmospheric CO_2. Figure 1 shows how dust transport to the oceans could conceivably modify ocean productivity and atmospheric CO_2 and the consequent impacts on other climate variables including the dust cycle. One major objective of palaeoclimatology is to use the dust record in ice cores and sediments, coupled with our knowledge of Earth surface processes today, to improve our understanding of these linkages in the current climate state and to use this information to better interpret the Earth's past climate history.

For the future, dust cycle models predict large changes in aeolian transport from the continents to the oceans over coming centuries, in response to anthropogenic climate change (e.g. Mahowald et al., 2006). In order to anticipate the effects of future changes in dust emissions, it is important to identify the impacts of past and present-day dust flux changes in terms of feedbacks with regard to radiative forcing of dust aerosols and any ocean fertilization and resultant productivity-driven changes in atmospheric CO_2.

Here, in order to examine past fluxes and possible climate impacts of dust in general and iron-bearing dust in particular, we synthesise available records of dust deposition at the last glacial maximum (LGM); evaluate the evidence for changes in ocean palaeo-productivity associated with, and possibly caused by, changes in aeolian flux to the oceans at the LGM; and consider the radiative forcing effects of increased LGM dust loadings. We begin by presenting an overview of global dust processes and the state of our knowledge about these processes.

Figure 1

2. Present-Day Global Dust Processes and Distributions

2.1 Dust source processes.

Dust mobilization is a highly complex process that is a function of many atmospheric, soil, and terrain properties (Goudie, 2008). Consequently the detachment, entrainment and transport of dust are highly variable both spatially and temporally. Dust generation is mostly associated with strong and/or gusty winds with the emitted flux varying as the third or fourth power of wind speed. The necessary wind fields to uplift dust can be generated by a wide range of local meteorological conditions. In particular, surface heat fluxes that induce convective activity can lead to dust generation (Engelstaedter and Washington, 2007), e.g., haboobs (Williams, 2008). Terrain features can strongly modulate emission rates (Washington et al., 2005; Koren et al., 2007). Particle detachment is inhibited by any element which increases surface roughness, e.g. vegetation, snow cover and surface armouring (by pebbles, stones or by salt crusts). Soil moisture plays a critical role by affecting particle cohesion along with cementation by salt content and soil structure. Dust emission thus tends to be favoured in arid or semi-arid areas, with annual rainfall < ~ 250 mm p.a. (Prospero et al., 2002; Washington et al., 2003), little or no vegetation cover and strong, long-fetch winds.

Many of the most intense present-day dust sources are associated with deep alluvial deposits laid down in the Pleistocene and Holocene (Prospero et al., 2002). Direct detachment and entrainment by the wind of particles (especially fine particles, < 20 μm, PM_{20}) appears subordinate to the effects of saltation (fig. 2); the impacts of sand-sized grains and/or aggregates on their downward trajectory cause 'sandblasting' - the emission of finer dust grains from the surface and/or from the original saltating aggregate (e.g. Gillette, 1978; Shao et al., 1993; Sow et

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al., 2009). Thus the dust mobilization process and the resulting properties of the deflated dust can be extremely sensitive to a wide range of environmental variables (including, for example, land use change (Tegen et al., 1995)). The complexity of these processes potentially makes dust a good proxy for climate variability but also makes the dust record difficult to interpret.

Figure 2.

2.2. Global dust sources and transport.

Much of our knowledge of the large-scale transports of dust is based on remotely sensed data, primarily from satellites. While there is an extensive literature of the directly measured concentrations and physical properties of dust, these are mostly too isolated in time and space to provide a coherent picture of dust transports. However, such measurements, when coupled with remote sensing data, can provide a compelling and informative picture of global dust transport. Satellites have come to play a dominant role in aerosol research (Kaufman et al., 2002). At present there is a large variety of sensors and platforms that provide information on aerosol concentrations and on some aspects of their physical properties - e.g., size, radiative absorption, polarization (Dubovik et al, 2008) - and also on the altitude distribution of these properties (e.g., Liu et al., 2008).

Satellite measurements of aerosol optical depth (AOD) with NOAA Advanced Very High Resolution Radiometer (AVHRR) provide a global picture of aerosol transport over the oceans (Fig. 3a; Husar et al., 1997). Over land, the most active dust source regions are detected with the Total Ozone Mapping Spectrometer (TOMS) absorbing aerosol product (Fig. 3b; Prospero et al., 2002). More recently, Shepanski et al. (2007) identified the regions where dust source is activated using three different infrared channels of the Meteosat Second Generation (MSG) instrument. This analysis is based on the frequency of dust generation but does not allow to retrieve the magnitude of the flux resulting from the episodes. Analyzing dust emission flux or evaluating dust mass requires a careful analysis of dust optical depth retrieved over deserts by the MISR (Multiangle Imaging SpectroRadiometer) instrument ,together with information on the vertical distribution of dust retrieved from CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation). Shepanski et al. (2009) were able to retrieve information on

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the diurnal cycle of dust emissions over Western Africa. Sixty five percents of dust source activation was found to occur in early morning from 0600–0900 UTC, well before the hours when the surface latent heat fluxes become important.

It is clear from Figure 3 that at the present day the northern hemisphere is far more 'dusty' than the southern hemisphere. This statement is based largely on *in situ* aerosol measurements made in different ocean regions (e.g. Duce et al., 1991; Prospero, 1996a; Prospero et al., 1989) and on satellite products such as shown in Figure 3. Satellite AOD distributions (fig. 3a) show large areas of high AOD over the oceans; the distribution of these plume-like features is clearly linked to continental sources. The largest and most persistent plumes (fig. 3a) emanate from arid regions in North Africa, the Middle East and Asia; the aerosols in these plume regions typically contain high concentrations of dust (e.g., Husar et al., 1997; Duce et al., 1991; Prospero, 1996; Prospero et al., 1989). These plumes can be directly linked to dust sources on the continents using the TOMS satellite (fig. 3b), one of the few sensors that can effectively sense aerosols over land (Herman et al., 1997).

It is notable that the major source regions (Fig. 3a and b; Table 1) are concentrated within a broad 'dust belt' that extends from the west coast of North Africa eastward to the Pacific coast of China. The timing of maximum dust emissions varies across this dust belt. In North Africa, maximum dust transport occurs in boreal summer, with dust transported across the Mediterranean to Europe and the Middle East (Moulin et al., 1998) and across the Atlantic to the Caribbean (Prospero and Lamb, 2003), the southeastern United States (Perry et al., 1997; Prospero, 1999), and the midlatitude western North Atlantic (Arimoto et al., 1995). There is also considerable transport in the winter with dust carried into South America (Prospero, 1981). In the Middle East, activity peaks in the late spring/summer and is at a minimum in the winter. Over the Indian subcontinent, activity peaks in the spring and decreases in the summer with the onset of the southwest monsoon. In Asia, dust activity peaks strongly in the boreal spring.

Of the major dust sources, North Africa is by far the strongest, contributing ~ 40-60 % of global dust emissions (e.g. Prospero, 1996b; Ginoux, 2001). For example, the so-called Sahara–Sahel 'dust corridor' extends over ~ 4000 km from Chad to Mauritania and emits very large volumes

of dust over the Atlantic Ocean (Moreno et al., 2006). Within this broad 'corridor', specific sources stand out as 'hot spots' of activity. Most notable is the Bodele Depression, Chad, the world's most persistently intense dust source at present day, responsible for an estimated ~ 15-20 % of North African emissions (Washington et al., 2006). While the Bodele stands out as a unique source on a global scale, much of North Africa serves as an effective dust source. Indeed, much of North Africa is frequently covered by a dust pall during much of the year. A large region of intense and frequent dust activity lies in Mali, Mauritania and southern Algeria (Prospero et al., 2002; Goudie and Middleton, 2006). Many of the most active sources are associated with drainage systems from the Ahaggar Mountains. This region is probably more representative generally of arid region dust sources and source processes, with many types of sources contributing to the dust emissions: i.e., old alluvial deposits, playas, dried lakes and wadis. Another area of dust activity lies in Tunisia and Northeast Algeria, greatest activity occurring in association with an extensive system of salt lakes and dry lakes in the lowlands south of the Tell Atlas Mountains. Major dust activity also occurs in the eastern Libyan desert in a region marked with an interconnected a chain of wadis (Wadi al-Farigh, Wadi al Hamim). Other active sites are found in Egypt, Sudan and the flanks of the Ethiopian Highlands.

The other major source regions of the Northern Hemisphere are the Arabian peninsula (~10-20 % of global emissions, Table 1) and central and eastern Asia (~15-20 %). For the Arabian peninsula, two major active areas extend along the eastern side of the peninsula along the Persian Gulf, and along the Oman coastal margins (fig. 3), comprising coastal sabkhas and dissected wadi systems, respectively (Goudie and Middleton, 2006). The lower Tigris-Euphrates basin, from north of Baghdad to the Persian Gulf, also provides a summer peak in dust activity, sourced from an immense alluvial plain. Very high concentrations of dust and pollution-derived species are found over the Arabian Sea early in the year (fig. 3a) carried by the winter monsoon and the action of systems moving from the North (Verma et al., 2007). Dust is carried from sources in the eastern Sahara and in northwest India (including the Thar Desert) and Pakistan. Concentrations of dust and pollutant species drop sharply across the inter-tropical convergence zone and with the onset of the southwest (summer) monsoon (Savoie et al., 1987; Krishnamurti et al., 1998) and demonstrate the effectiveness of the inter-tropical convergence zone in blocking the transport of aerosols to the southern hemisphere.

In eastern Asia, dust sources - mostly in China - are extremely active in the boreal spring. Very large quantities of dust (often mixed with high concentrations of pollutants) are carried out of China eastward, across the coastal waters, and over Korea and Japan where the dust is such a persistent phenomenon, noted in many centuries of historical records, that it is given a name, 'kosa' – yellow dust (Kar and Takeuchi, 2004). Asian dust is routinely observed in relatively high concentrations at stations in the central North Pacific (Prospero et al., 1989). The strong seasonal dust cycle and the trans-Pacific extent of the transport can be seen in figure 3a. Although there are many active dust sources in China (Prospero et al., 2002; Goudie and Middleton, 2006; Wang et al., 2004), several stand out in satellite images: the Tarim Pendi basin (containing the Taklimakan Desert; central Inner Mongolia Plateau and the Gobi Desert); the Hexi Corridor and the Loess Plateau; and western Inner Mongolian Plateau. The Tarim Pendi basin, which receives very little rainfall (50-100 mm a year along its periphery and ~ 10 mm in the central regions), is most active from February through May. The Gobi desert, located east of the Tarim Basin on the Mongolian Plateau, is a major dust source at present. Every spring, spectacular dust storms can be seen generated over the Gobi; similar to many dust events in North Africa, the most active sources cover a relatively small area of the surface in effect acting as 'point' sources. Gobi dust outbreaks can be followed n satellite imagery as they move across the Pacific to North America (Husar et al., 2001). The Loess Plateau extends over ~440,000 km^2 and is covered with loess with depths ranging from typically ~100 m to as much as 350 m (Maher et al., 2008). The Loess Plateau is unique in the global dust picture in that it is the only loess region that serves as a highly active dust source at the present day. These loessic soils are now susceptible to enhanced deflation because of the intense agricultural activity in this region (Wang et al., 2004). While various estimates exist as to the quantitative effects of human activities in dust mobilization (*see Section 1)*, it is clear that of all the major dust sources, the Asian sources are those where humans could be having a major effect on emissions, although this is still a matter of considerable debate (Wang et al., 2006).

Figure 3 a and b

For the southern hemisphere, contemporary dust sources have been mapped by Prospero et al. (2002) using TOMS data. Australia, South America and southern Africa each have localized areas of dust emissions. However, there is little evidence of major and sustained dust activity in these regions. Although there are few measurements of aerosols over the southern oceans, these show much lower concentrations than over the northern oceans (e.g., Prospero et al., 1989; Baker et al., 2006; Wagener et al., 2008). Measurements of AOD (e.g. Fig. 3a) made by a wide variety of satellite systems (Zhu et al., 2007) fail to detect substantial aerosol plume effects that can be attributed to dust sources. This may partly reflect some under-estimation by TOMS due to difficulties of dust detection in cloudy conditions (e.g. Gassó and Stein, 2007). However, compilations of satellite aerosol event imagery contain many hundreds of examples of dust plumes, the overwhelming majority of which are linked to Northern Hemisphere sources (see, for example: http://visibleearth.nasa.gov/; http://rapidfire.sci.gsfc.nasa.gov/gallery/). In stark contrast, model results (IPCC, 2001a; Li et al., 2008) often identify emission of large dust plumes from Australia (comparable to those sourced from NW Africa). Ground-truthing of either the remotely sensed or modelled data is severely constrained by the lack of modern dust data. For example, while the ModelE dust component of the GISS Global Circulation Model (Cakmur et al., 2006) is constrained by data from 96 individual sites, only 18 of those are in the southern hemisphere (and of these, 13 were measured during a historic low for dust emissions from Australia (Mackie et al., 2008). The main Australian source is the Lake Eyre basin (Bullard et al., 2008), followed by the Murray-Darling basin, both comprising extensive river systems, floodplains and dunefields (Bullard and McTainsh, 2003). There are two main dust transport paths, one to the northwest and one to the southeast (Bowler, 1976). Although the southeast path is often described as crossing the Tasman Sea (and Australian dust is often reported in New Zealand, e.g. Marx et al., 2005), individual events are variable and can contribute sediment further south, to the Southern Ocean and southwest Pacific (McGowan et al., 2000; McTainsh et al., 2005; Shao et al., 2007). Iron is a conspicuous component of Australian soils and dust storms (Taylor et al., 1983; McTainsh, 1989; Mackie et al., 2008) but the concentrations and mineralogy of Antipodean long range transport (LRT) dusts are poorly known. From magnetic measurements, modern dusts from eastern Australia and the palaeo-dust contained within Tasman Sea sediments contain fine-grained haematite/goethite, which varies directly with the concentration of the mineral dust fraction (Hesse, 1997). New Zealand appears to be an

301 insignificant source of dust at the present day (Prospero et al, 2002), unsurprising as prevailing
302 trans-Tasman winds are to the east (McGowan et al., 2000).
303
304 Although southern Africa has some significant dust sources (notably the Etosha, Namibia and
305 Magkadikgadi, NE Botswana, pans; Prospero et al., 1992; Washington et al. 2003), it is not a
306 major exporter of dust. Iron-rich South African dust that enters the ocean is largely (> 90 %)
307 deposited in the S. Indian Ocean (e.g. Piketh et al., 2000) along latitude ~ 35 °E with only ~ 4 %
308 entering the S. Atlantic (Garstang et al., 1996) Some work (Stuut et al. 2005) points to deflation
309 of silt-rich (> 20 μm) dust to the southwest into the South Atlantic, but it is unknown if this is
310 then transported into the higher latitudes.
311 For South America, the southern region is spanned by a continuous band of arid or semi-arid
312 terrains extending from the Peruvian to the Patagonian coast (the 'Diagonal àrida'/*arid*
313 *diagonal*). Based on TOMS data, Prospero et al. (2002) identified three main dust sources in
314 southern S. America which overlap most of this *arid diagonal*: the Puna (22-26 °S)/Altiplano
315 (15-22 °S) Plateau, central-west Argentina (27-35 °S) and Patagonia (39-52 °S). Preferential
316 emission sources appear linked to large dry lakes (Puna/Altiplano), thousands of small enclosed
317 basins (Patagonia) and extensive alluvial fans (central-west Argentina). Patagonia is strongly
318 influenced by the southern hemisphere westerlies. Patagonian-sourced dust is estimated to supply
319 ~90 % (~ 30 g m^{-2} a^{-1}) of the present day sediment flux to the SW Atlantic from S. America
320 (Gaiero et al. 2003). In the high arid plateau (Puna/Altiplano) of the central Andes, the climate is
321 mostly governed by the subtropical jet stream and dust emitted from this area can be directly
322 injected into the tropospheric winds. Both wind systems can produce eastward dust transport and
323 deposition to proximal areas like the Pampas and the nearby Atlantic Ocean or, distally, to the
324 Southern Ocean or the East Antarctic Plateau. The characteristics of present day Patagonian dust
325 are relatively well known (Gaiero et al., 2003; Gaiero et al., 2004; Gaiero et al., 2007). Clay-size
326 fractions are dominated by the clay mineral, smectite. The chemical/isotopic composition of dust
327 reflects the contribution of Jurassic and Quaternary explosive volcanism (Gaiero et al., 2007).
328 Dust deposition rates vary from ~5 g m^{-2} a^{-1} at the Patagonian coast, ~28 g m^{-2} a^{-1} (Gaiero et al.,
329 in prep.) at the Central Pampa (33 °S); and ~26 g m^{-2} a^{-1} for the southern Pampa (39 °S) regions
330 (Ramsperberger et al., 1998; Gaiero et al., 2003).
331

Major dust sources require not only the operation of efficient dust deflation and transport processes but also a replenishment of dust supply, through active weathering and comminution of parent materials. TOMS (fig. 3b) shows a clear association of dust sources with specific types of terrains and environments. All major sources in figure 2b are located in arid regions (annual rainfall < ~ 200–250 mm) and centered over topographical lows or on lands adjacent to strong topographical highs. Although the source regions themselves are arid or hyper-arid, fluvial action is evident everywhere by the presence of ephemeral rivers and streams, alluvial fans, playas, and saline lakes. Furthermore, even presently arid source areas have had a relatively recent pluvial history. The association of dust sources with water features is consistent with our understanding of the processes involved in the production of fine particles through weathering (Pye, 1989; Smalley et al., 2005). Fluvial and chemical weathering processes appear much more efficient in the production of small particles (i.e., particles < ~10 μm diameter) than are aeolian processes (i.e. grinding and impaction). Subsequently, through fluvial action, small particles are carried to depositional basins or alluvial plains where, after drying, they are subject to deflation by wind. Thus, precipitation totals and timing are important both in the (low-lying) area of emission and in any adjacent, dust-supplying highlands. TOMS shows a remarkably consistent association of dust sources with playas and associated terrain environments [Prospero et al., 2002], such as the alluvial fans that ring the basins in which the playas are found (Reheis et al., 1995). The playa is the ultimate receptacle of almost all the fine-grained sediments eroded within a basin over a geological time period, thus they are storehouses of fine, dry, unconsolidated (and thus wind-erodible) sediments often in amounts far in excess of production rates in the present climate. An important characteristic of other major dust sources is the presence of deep and extensive alluvial deposits. During pluvial phases, these basins were flooded and thick layers of sediment were deposited which are now exposed. Many of the most active TOMS sources were flooded during the Pleistocene - Holocene. The prime example is the Bodele Depression in Chad, the largest source of long-range dust in the world (Warren et al., 2007).

Thus, the most productive dust-source environments at the present day are the result of the complex interplay between climate, terrain and geological history. In order to understand the linkages between climate, dust production, and transport, we must understand in detail how these features interact.

Over the past decade, global circulation models and chemical transport models have come to play an important role in understanding dust-climate relationships. At present, there are over a dozen models that simulate the dust cycle, represent its optical properties and predict dry and wet deposition over the global domain (Textor et al., 2006). However, the accuracy of global aerosol models is limited by uncertainties in aerosol emission source characteristics, in our knowledge of atmospheric transport and removal processes and also errors in the meteorological data assimilated into the model computations. Thus there can be large differences among the outputs of the various models and, in the absence of aerosol measurements over large areas of the Earth, it is difficult to select the "best" model. Regardless of these shortcomings, most of the currently reported dust budget values are based on transport models (e.g. Werner et al., 2002; Luo et al., 2003; Zender et al., 2003 and 2004; Ginoux et al., 2004; Mahowald et al., 2005; Cakmur et al., 2006; Miller et al., 2006; Tanaka and Chiba, 2006; Li et al., 2008).

3. Dust impacts on climate.

3.1 Dust as a source of bioavailable iron

Dust may play a critical, indirect role in climate change, through biogeochemical interactions (e.g. via iron or other essential element fertilization of ocean and/or terrestrial ecosystems) and chemical interactions with ozone and sulphur cycles (Cwiertny et al., 2008; Chin et al., 2000). In the case of ocean fertilization, dust containing iron, as fine-grained iron oxides/oxyhydroxides and/or within clay minerals, and other essential micronutrients (e.g. silica) may play a significant role for those regions of the world's oceans where macronutrients like nitrate are abundant but primary production and nitrogen fixation is limited by iron scarcity. Hence, as formalised by Martin et al. (1990, 1991), the efficiency of the 'biological pump' is low and there is significant potential for enhanced export production and drawdown of atmospheric CO_2. The Southern Ocean and the equatorial and northwestern subarctic Pacific are the major high nitrate, low chlorophyll (HNLC) areas, totalling some 30-50 % of the world ocean (Moore et al., 2004; Aumont et al., 2003). Diatoms are the key phytoplankton group for formation of algal blooms in such areas and are strongly iron-limited. Additionally, in tropical and subtropical regions, iron deficiency may also limit phytoplankton growth in subtropical gyres (Johnson et al., 1997; Sedwick et al., 2005) and nitrogen fixation by the diazotrophs (Falkowski, 1997; Moore et al., 2006). Supply of iron-bearing dust may thus directly and indirectly limit primary productivity for

394 large portions of the world ocean (Moore and Doney, 2007), affecting biological carbon export at
395 the global scale (Moore and Braucher, 2008).
396
397 Consistently higher dissolved iron concentrations (>1 nM) are observed for ocean areas below
398 major dust plumes (Sarthou et al., 2003). The 'iron hypothesis' (Martin, 1990) suggests that past
399 increases in aeolian, iron-bearing dust supply to the surface of HNLC ocean waters during
400 glacial stages relieved iron limitation on primary productivity. The resultant increase in the
401 export of biogenic carbon from surface to deep ocean waters (the 'biological pump') caused a
402 decrease in surface ocean pCO_2 that was balanced by an increased drawdown of CO_2 from the
403 atmosphere. The details of this process, and the possible magnitude of its effects on the global
404 carbon cycle, are the subject of much international debate (e.g. Honda et al., 2006; Maher and
405 Dennis, 2001; Moore and Doney, 2007).
406
407 The sensitivity of HNLC regions to increased iron supply has been demonstrated by a series of
408 artificial mesoscale iron fertilization experiments in the HNLC regions (e.g. Boyd et al., 2007;
409 Coale et al., 2004; Coale et al., 1996). The common outcomes to these experiments were the
410 expansion of phytoplankton standing stocks, with distinct population shifts to large diatom
411 species, and resultant drawdown in atmospheric CO_2 (deBaar et al., 2008; Tsuda et al., 2003). It
412 has been difficult to quantify export production via subsurface storage of carbon (rather than
413 subsequent re-release from the ocean mixed layer). For the NW Pacific SEEDS experiment,
414 drawdown of atmospheric carbon was estimated at ~ 150-1500 C atoms per iron atom added
415 (Tsuda et al., 2003). The most recent experiment was also the largest (Lohafex, 2009), with 6
416 tonnes of dissolved iron added to a 300 km^2 ocean patch in the southwest Atlantic sector of the
417 Southern Ocean. However, whilst there was a doubling of (non-diatom) phytoplankton biomass
418 within the first two weeks, increased grazing pressure of zooplankton prevented further
419 development of the algal bloom, with only minor amounts of carbon being removed from the
420 ocean surface layer as a result.
421
422 Based on these magnitudes of export production, and taking into account the rather low fractions
423 of iron estimated to be bioavailable from mineral dust (e.g. Baker et al., 2006a), modelled
424 glacial-stage changes in atmospheric CO_2 resulting from dust-driven iron fertilisation range from

425 ~ 20 % (Bopp et al., 2003) to > 50 % (Watson et al., 2000) of the total interglacial-glacial change
426 of ~ 100 ppmv. However, extrapolation of the artificial iron enrichment experiments to estimate
427 past carbon sequestration may be unrealistic given their short duration and use of unfeasibly
428 large and rapid iron additions, which lead to loss of most (80 – 95 %) of the iron added during
429 the enrichment experiments (Bowie et al., 2001). More realistic estimates of carbon sequestration
430 may be derived from analysis of a recent (Nov. 2004 – Feb. 2005), natural phytoplankton bloom
431 in the Southern Ocean (Blain et al., 2007). This natural event was driven by indirect iron supply
432 from upwelling of bottom waters (Lefevre and Watson, 1999; Maher and Dennis, 2001), rather
433 than direct aeolian iron supply. pCO_2 measurements of the surface waters showed that the bloom
434 was an important CO_2 sink, with mean pCO_2 drawdown 2–3 x higher than that observed for the
435 artificial Southern Ocean iron experiments, probably reflecting the longer duration of the natural
436 bloom. Carbon sequestration efficiency was estimated to be at least 10 times higher than that
437 estimated from the artificial experiments. The higher sequestration efficiency of the natural
438 bloom has been attributed to its more complete and longer-lasting development, aided by slow
439 and continuous natural additions of iron (and eventually other required macro-nutrients), from
440 adjacent deep water supply (Blain et al., 2007). These results indicate first that aeolian supply of
441 iron to the oceans, both directly and indirectly (i.e. deposited elsewhere and subsequently
442 dissolved and transported), may have been significant in palaeo-ocean and atmospheric CO_2
443 changes. Second, the HNLC oceans may presently be, and previously have been, more sensitive
444 to aeolian iron supply than has been indicated to date by the artificial experiments. However,
445 the SOIREE experiment (Boyd and Abraham, 2001) showed that the photosynthetic competency
446 of iron-limited Southern Ocean phytoplankton increased only when the concentration of
447 dissolved iron exceeded a threshold value of ~0.2 nM, i.e. ~ 2 x twice the ambient $[Fe]_{dissolved}$
448 value. Deposition of dust can thus increase the iron inventory of surface waters without
449 stimulating a phytoplankton bloom (Boyd et al., 2009). Previous reports of correlation between
450 dust events and remotely observed increases in surface water chlorophyll (e.g. Gabric et al.,
451 2002; Shaw et al., 2008) need to be evaluated with respect to a physiologically realistic lag time
452 between the increase in dust/iron supply and phytoplankton response (Boyd et al., 2004). For
453 example, whilst Shaw et al. (2008) attributed a bloom in waters of the Great Barrier Reef Lagoon
454 (<50 km from mainland) coastal to a large dust storm (the largest in over 40 years), Mackie
455 (2009) pointed out that the time (<<1 day) between dust deposition and observations of

456 chlorophyll increase precluded dust as a causal agent. In response, Shaw et al. (2009) suggested
457 that smaller dust events before the large dust storm were responsible. However, in waters so
458 close to the coast, subject to numerous such small dust events, it is unlikely that iron limitation
459 would develop in any case.
460
461 An assumption of many of the dust-cycle and iron supply models is that atmospheric input of
462 bioavailable iron to the surface ocean varies linearly with dust loading. Present day field
463 collection and dust leaching studies (Sedwick et al., 2007; Baker & Jickells, 2006; Jickells et al.,
464 2005) indicate that it is critical to identify the nature (mineralogy), source (e.g. size distribution)
465 and transport history of the dust, in order to evaluate the possible proportion of bioavailable iron
466 carried within the terrigenous dust loading (Baker and Jickells, 2006; Baker and Croot, 2009). A
467 diverse range of dissolution experiments has been reported for a range of iron-bearing
468 aluminosilicates and iron oxides (e.g. Journet et al., 2008), potential source soils (e.g. Visser et
469 al., 2003) and increasing numbers of natural dust samples (e.g. Spokes et al., 1994; Baker et al.,
470 2006b; Desboeufs et al., 2005; Mackie et al., 2005). Generally low but spatially variable iron
471 solubilities appear the norm, i.e. < 0.6 % (e.g. Baker et al., 2006b; Journet et al., 2008). At or
472 close to source areas, soil mineralogy appears to be the key influence on iron solubility. For
473 example, for some African sources, higher solubilities (0.3 %) were displayed by iron-bearing,
474 smectite-rich dust from the central Sahara and lower values (0.1 %) by illite/haematite/goethite-
475 rich dust from the western Sahara and the Sahel (Journet et al., 2008). Conversely, at areas
476 remote from major dust sources, and associated with decreased dust concentrations, increased
477 levels of iron solubility have been reported, and ascribed to progressive particle fining with long
478 range transport (e.g. Baker and Croot, 2009). However, as discussed below, the particle size of
479 LRT dust appears to stabilise rather than become progressively finer. Other factors of likely
480 significance for iron solubility include enhancement of fine particle reactivity during the
481 transport process, especially related to ligand formation and surface adsorption and/or desorption
482 processes. Depending on ambient pH, iron-hydroxyl groups at iron oxide surfaces can achieve
483 negative surface charge by dissociation ($Fe\text{-}OH \rightarrow FeO^- + H^+$) or association ($FeOH + H^+ \rightarrow FeOH^{2+}$) of protons (e.g. Cornell and Schwertmann, 2003; Taylor et al., 1983). For the iron
484
485 oxides/ hydroxides, the zero point of charge (zpc, i.e. where there are equal concentrations of
486 surface FeO^- and $FeOH^{2+}$) varies between ~ pH 5 - 8. Similar pH-dependence of surface and

inter-layer charge is displayed by the clay minerals, but with much lower zpc values, from < pH 2-3 for smectite to pH 2 – 4.5 for kaolinite . Below the zpc, negatively-charged adsorbates, such as natural organic matter, can subsequently be retained at or near the mineral surface. Such adsorption may play a key role both in reducing particle scavenging and increasing iron solubility in the marine environment. Iron reduction can result from photolysis of Fe^{3+} - complexed with bacterially-produced siderophores (Barbeau et al., 2001) complexes. The solubilised Fe^{2+} could then undergo direct biological uptake, be re-oxidised to Fe^{3+} and/or subjected to subsequent complexation. Recent reports suggest that Fe-reducing compounds, such as dimethyl sulphide, might interactively be released from iron-deficient phytoplankton (Johansen and Key, 2006), although the iron compound modelled in this study was ferrihydrite, the most microcrystalline and reactive of the soil oxyhydroxides. It is clear that large uncertainties presently remain regarding the biogeochemistry of iron complexation, release, uptake and scavenging in the marine environment (Baker and Croot, 2009).

The interpretation of Fe solubility in aerosol samples is complicated at the present day by the possible presence of Fe derived from anthropogenic sources, in particular, combustion processes, both industrial and biomass burning. Measurements on Bermuda (Sedwick et al., 2007) show that air masses from North American air typically contain relatively low concentration of aerosol iron (0.5 nmol Fe m^{-3}) but the fractional solubility is very high, 19 %. In contrast dust-laden African air masses contained relatively high concentrations of aerosol iron (27.8 nmol Fe m^{-3}) but the fractional solubility was low, only 0.44 %. The net result is that the quantity of soluble Fe derived from the aerosols was the same in both cases. The finest particle size fraction yielded the highest solubility for both aerosol types. Model results (Luo et al., 2008) suggest that combustion-derived iron can account for as much as 50 % of the total iron deposited in some regions. However, over the open ocean it is usually less than 5 % of the total iron although close to the East Asian continent in the North Pacific values can be as high as 30 %.

The observation of increased iron solubility associated with low dust concentrations would imply proportionally enhanced supply of bioavailable iron to ocean areas distal from dust sources (notably, the Southern Ocean) and less to high dust flux areas (Baker and Croot, 2009). The nature of the causal link between dust concentration and solubility requires further evaluation, in

518 order for robust incorporation of varying values of iron solubility of dust into global models of
519 iron flux (e.g. Fung et al., 2000; Bopp et al., 2003). Most models for dust-iron processes use a
520 single uniform value for iron solubility from dust but a recent report of 2-3 % solubility of iron
521 from glacial flour compared to <1 % for arid soils (Schroth et al., 2009) provides a new variable
522 for models to incorporate. The impacts of ocean acidification, likely to modify aerosol and iron
523 solubilisation processes, also require evaluation (Breitbarth et al., 2009).
524
525 *3.2 Dust impacts on climate: radiative effects.*
526 *3.2.1 Dust particle size*
527 Dust size distributions in ocean sediments and ice cores are often used as a measure of dust
528 generation and transport dynamics. The size distributions of dust particles also have a critical
529 influence on the direct and indirect climate impact of dust (see below) and they are also a major
530 uncertainty in dust modelling. Most of the long-traveled dust mass is located in the size range
531 under about 20 μm diameter (e.g. Arimoto et al., 1997), with a mass median diameter of ~ 1.5 –
532 3 μm (Balkanski et al., 1995; Reid et al., 2003). Particles in this size range can remain suspended
533 in the troposphere for days to weeks, and be carried over great distances (>> 1000 km) before
534 being removed from the atmosphere by deposition processes. Asian dust, for example, has been
535 shown to contribute to dust samples spanning western to eastern China (Gao et al., 1992), the
536 Yellow Sea (Gao et al., 1997), Japan and Korea (e.g. Park et al., 2005), the Pacific Ocean (e.g.
537 Duce et al., 1980; Rea et al., 1985; Uematsu et al., 1985). More distally, it has a significant
538 impact on air quality in the western United States (VanCurren and Cahill, 2002); and is a major
539 source of dust in Greenland ice cores (Bory et al., 2003). North African dust is carried in great
540 quantities to the Amazon basin (Swap et al., 1992), into the Caribbean (Prospero and Lamb,
541 2003) and the southeastern (Prospero, 1999), eastern and northeastern US (Perry et al., 1997).
542 The first challenge for improving data on dust particle size distributions, whether at present day
543 or for the palaeo-record, is the need for expanded geographic and temporal coverage. The second
544 requires instrumental convergence. Across the modern and palaeo-data aerosol communities,
545 particle size tends to be measured using different instruments. For ice and sediment cores, for
546 example, particle size analyses are most frequently derived from instruments using the Coulter
547 Principle (such as the Coulter Multisizer). In contrast, the atmospheric community uses a wide
548 range of instruments that are based on different physical principles, i.e., the mobility of aerosols

549 in electric fields, the optical properties of aerosols, aerosol aerodynamic properties. These
550 instruments yield widely disparate results even when making measurements on identical dust
551 aerosol populations, including African dust (e.g. Reid et al., 2003).
552
553 Notwithstanding this instrumental caveat, modern aerosol size data suggest evolution of the
554 modal diameters for long range transport (LRT, i.e. > 1000 km) dust to a rather invariant range
555 of ~ 3 to 3.5 μm diameter, in contrast to values obtained in or near source regions, which tend to
556 cluster at ~ 5 μm diameter (Sviridenkov et al., 1993; Arimoto et al., 1997, Moore et al., 2004;
557 Maring et al., 2003). For example, dust measured on the Canary Islands (300 km from the N.
558 African coast) and in Puerto Rico (5000 km to the west) shows very similar particle size
559 distributions for the < 7 μm size range (fig. 4a). Larger particles (> ~ 7-8 μm diameter) do
560 display between-event differences, due to their preferential removal during atmospheric
561 transport.
562
563 Similarly, in surface sediments along a 3600 km-long, west-east transect of the North Pacific, a
564 transect lying ~ 4000-8000 km downwind from the E. Asian source areas (i.e. distal from any
565 possible non-aeolian terrigenous sources), the median dust grain size barely changes, at ~ 2.8 μm
566 to 2.4 μm (Janecek, 1985). More recent atmospheric sampling campaigns also suggest a size
567 range of ~ 1- 3 μm for LRT particles in this region (e.g. Zhao et al., 2003). A simple empirical
568 model, which sets the vertical velocity of dust particles equal to the Stokes gravitational settling
569 velocity minus an upward velocity of ~0.33 cm s^{-1}, accurately predicts changes in dust size
570 distribution during atmospheric transport (Maring et al., 2003). Thus it appears that some
571 atmospheric process or processes partially counteract gravitational settling along long transport
572 pathways. It should also be noted that the transport of giant, >75 μm, grains of dust for up to
573 10,000 km has been reported (e.g. Pitty, 1968; Betzer et al., 1988, Middleton et al., 2001) and
574 can not be explained in terms of simple Stokes type settling. The presence of long-lived, long-
575 travelled dust concentrations within the atmosphere remains an unresolved challenge to our
576 understanding of LRT processes.
577
578 Such relative observed invariance of dust size also suggests some degree of constancy in dust
579 mineralogy, morphology and size at the regional scale (e.g. fig. 4a), in turn indicating that source

580 soils or sediments are a factor in controlling dust particle size, in addition to wind speed and/or
581 in-transport processing (Grini and Zender, 2004). In this scenario, dust properties remain
582 consistent when the sources also remain constant, but can be expected to change if and when the
583 source areas change. Wang et al. (2007) report similar conclusions for modern dusts sourced
584 from NW China. The dust grain size distributions vary from source to source but are generally
585 trimodal, with the first peak for particles > 11 μm, the second for particles 4.7 – 7 μm, the third
586 for sub-micrometre particles (< 0.43 μm). These distributions remain essentially the same for any
587 one source whether under dust storm or non-dust storm conditions. As noted by Wake and
588 Mayewski (1994), distinctive variations have been observed in the particle number- and mass-
589 size distributions of dusts collected from different geographic regions, including Europe (e.g.
590 Junge, 1963), the Sahara (Schutz and Jaenicke, 1974), the Swiss Alps (Wagenbach and Geis,
591 1987), Colorado and Texas (Patterson and Gillette, 1976), Tajikistan (Gomes and Gillette, 1993)
592 and Greenland (Steffensen, 1985, 1997).
593 These observational data do not support the wind speed dependence of LRT dust particle size
594 (fig. 4b) reported experimentally by Alfaro et al. (1998) and by observation by Sow et al. (2009).
595 These studies indicate enhancement of very fine (< 2 μm) particles with increased wind speeds
596 and surface shear velocity (u*), as a result of 'sandblasting' by saltating grains. *Within* an
597 erosional event, however, the dust size distribution appears relatively insensitive to measured
598 changes of u*. This apparent conflict may reflect a dissonance between the typically long u*
599 averaging time (15 mins) and significantly shorter response times of the dust emission processes
600 (Sow et al., 2009). Engelstaedter et al. (2007), in contrast, use AERONET data to support the
601 conventional view that larger particle sizes reflect stronger, or possibly gustier, winds.
602
603 Figure 4.
604
605 *3.2.2 Dust radiative properties: effective particle size and mineralogy.*
606 In terms of radiative effects (RE), atmospheric dust can both scatter and absorb incoming solar
607 and outgoing long-wave radiation (direct RE), which alters the energy balance of the atmosphere
608 (IPCC 2001b; IPCC 2007), and can in turn produce changes in atmospheric circulation (Miller
609 and Tegen, 1998) and cloud cover (Perlwitz and Miller, 2009). The effect of dust absorptivity is
610 to produce warming of the atmosphere, locally or regionally, and cooling of the surface. The

611 effect of scattering is to produce more widespread cooling, the result of increased upwards
612 radiance. Dust also plays an important role in cloud microphysics, particularly with regard to
613 numbers of cloud condensation and ice nuclei, thus influencing the formation and lifetime of
614 clouds (indirect RE), which in turn influences the planetary albedo (e.g. Spracklen et al., 2008).
615 Estimates from dust-cycle models of the net top of atmosphere direct RE currently span both
616 negative and positive values, ranging from -0.6 to +0.4 Wm^{-2} (IPCC 2007). This uncertainty in
617 both sign and amplitude of dust radiative forcing at the present day reflects both the complexity
618 of dust distribution (including its vertical distribution, interaction with clouds, the albedo of the
619 underlying surface) and poor understanding of the scattering/absorptivity of dust particles, in
620 turn related to poorly known dust concentrations, mineralogy, particle size, and shape (e.g.
621 Nousiainen, 2009). These latter properties vary regionally, depending on the source materials
622 (soils, dry lake beds, ephemeral channels etc); and for any population of grains within a parcel of
623 entrained dust, they also vary interdependently. Mineral dusts typically consist of an internal
624 mixture of mineral species (and particle morphologies), often including quartz, clay minerals,
625 iron oxides and calcium carbonate (Claquin et al., 1999), each with their own optical properties
626 (Sokolik and Toon, 1999). Platy clay mineral particles, for example have different optical
627 properties than equidimensional silicates (Nousiainen et al., 2009). Recent estimates in radiative
628 forcing computations indicate that assuming that dust particles are homogenous spheres causes
629 roughly as large an error as that associated with the uncertainties in refractive index (Kahnert et
630 al., 2007). The latter has previously been considered the dominant source of uncertainty in dust
631 optical properties. The radiative behaviour of dust particles also depends on their effective
632 particle size, i.e. in relation to the wavelength under consideration. Thus, particles with diameters
633 of around 0.5 – 1 μm are effective in scattering solar radiation (fig. 5), relatively unaffected by
634 particle shape or surface roughness effects. Such effects become increasingly important for
635 wavelength-scale and larger particles (Nousiainen, 2009), including therefore those within the ~
636 1.5 - 3 μm size range (i.e. the reported mass median diameter for LRT dust, as above).
637
638 Figure 5
639 Another poorly quantified potential control on dust absorptivity is the presence of iron oxides/
640 oxyhydroxides, as so-called 'free iron', i.e. often as discrete, sub-micrometre particles (figure 6),
641 or as surface coatings on quartz grains, and, more rarely, iron oxide-clay aggregates (Sokolik and

642 Toon, 1999). Only where soil pH is low will iron oxide grains carry sufficient positive charge,
643 and/or the clay particles carry sufficient negative charge, to interact with each other (Cornell and
644 Schwertmann, 2003).
645
646 Figure 6.
647
648 Haematite and goethite occur as minor soil components (< ~ 5 % by mass) on a global scale,
649 reflecting the near-surface weathering of Fe^{2+}-containing primary (lithogenic) minerals by
650 hydrolysis and oxidation. Given that they occur in minor concentrations, both these minerals
651 have a disproportionate radiative impact, displaying large absorbing potential at shorter
652 wavelengths (Alfaro et al., 2004; Sokolik and Toon, 1999; Balkanski et al., 1997). The
653 mineralogy and concentration of iron oxides in dusts is dominantly controlled by the source soil
654 hydrology and mineralogy (Schwertmann and Taylor, 1977), subsequently by any erosional
655 processes of grain aggregation and/or disaggregation, and finally, by any particle size changes
656 during transport (e.g. Zhang et al., 2005). Establishing mineralogical linkages between potential
657 sources and airborne dust is likely to be most robust when based on particle size-specific
658 comparisons between the clay and fine silt fractions of source soils/ sediments and the fine and
659 coarse mode of sampled dust (Maher et al., 2009; Sokolik and Toon, 1999). Compared with its
660 'parent' soil, dust is likely to be biased towards smaller, less dense, and flatter (large surface-
661 area-to-volume ratio) particles (Nousiainen, 2009). Haematite and goethite tend to be associated
662 with fine (<< 10μm, i.e. fine silt and clay-sized) particles, with long residence times (i.e. days) in
663 the atmosphere and thus potentially long transport paths.
664 Haematite has been identified as a significant absorber in terms of dust radiative properties
665 (especially when present as sub-micrometre grains, fig. 6b and c). It is very stable, is the end
666 product of the dehydroxylation of all iron oxyhydroxides and is thus prevalent in warmer and
667 drier regions and increasingly limited in occurrence pole-wards of latitudes ~ 40 º N and S
668 (Cornell and Scherwtmann, 2003). For modern dusts from arid and semi-arid regions (Claquin et
669 al., 1999), haematite content varies between 0.6 to 3.4 % (by volume). The iron oxyhydroxide,
670 goethite, is less restricted in its distribution. Although the red coloration of many soils and dusts
671 often reflects the presence of haematite (highly pigmenting when it occurs as sub-micrometre
672 grains), goethite is often volumetrically the more important. In the loess and palaeosols of the

673 Chinese Loess Plateau, for example, the wt % of goethite is 2 –3 x higher that of haematite
674 (Balsam at al., 2007; Ji et al., 2004). Similar dominance of goethite has been reported for a
675 limited number of dust source samples from the Sahara and the Sahel (Lafon et al., 2006). The
676 presence of goethite can lead to large differences in the wavelength dependence of the single
677 scattering albedo (Sokolik and Toon, 1999). The evenness of distribution of goethite in the soil
678 matrix and resultant dusts is also likely to maximize its absorptivity. Inclusion of goethite in dust
679 modelling thus seems essential, given its near-ubiquity in source soils.
680
681 Direct RE forcing by dust appears to result in cooling over much of the Earth's surface (fig. 7,
682 and e.g. Perlwitz et al., 2001; Zhu et al., 2007). Conversely, over bright surfaces (such as
683 deserts), dust can exert significant warming effects. For example, discrepancies between
684 measured and modelled outgoing radiation over cloud-free, desert areas of north Africa indicate
685 that dust may exert a long wave radiation forcing as great as 50 Wm^{-2} in the monthly mean at
686 1200 U TC (Haywood et al., 2005). Similarly, for the 'great Indian desert' (Thar) area, warming
687 of the lower atmosphere from dust absorptivity (greatest during the winter season) has been
688 estimated at 0.7–1.2 K day^{-1} (Moorthy et al., 2007). Due to direct and indirect RE, dust can
689 regulate and interact with the heat sources and sinks of monsoonal regions, altering the
690 monsoonal water budget (Lau and Kim, 2006). A further complication arises from increased
691 emissions of anthropogenic, carbonaceous aerosols, which have reduced the global annual
692 average single scattering albedo; aerosol at present day is estimated to be approximately twice as
693 absorbing as that in preindustrial conditions (Myhre, 2009).
694
695 Given the potential impacts of effective particle size and iron mineralogy, improved modelling of
696 dust optical properties at solar wavelengths can only be achieved with improved data on present
697 and past regional variations in aerosol size and shape distributions, and in iron oxide
698 concentrations, mineralogies and mixing state. Given that haematite and goethite occur mostly in
699 minor concentrations, their quantification in dusts and sediments requires from any method of
700 analysis the capacity to analyse to low concentrations (i.e. below the detection levels of x ray
701 diffraction, for example). Diffuse reflectance spectrometry and environmental magnetic methods
702 offer potential in this regard, these measurements responding selectively to the iron oxide-

specific signals of colour and magnetic properties, respectively (Arimoto et al., 2002; Balsam et al., 2007; Maher et al., 2007; Watkins et al., 2007).

Figure 7.

4. *The palaeo-dust record for the last glacial maximum.*

4.1 Dust fluxes, size distributions, mineralogy.

Glacial/interglacial changes in the flux and particle size of dust have been recorded in ice cores and sediment records, both continental (windblown loess, lake sediment and peat records) and marine. Spanning the entire transect from pole to equator to pole, these dust records have shown that dust flux was higher in both hemispheres during past glacial stages (fig. 8). In light of the ongoing debates regarding modern dust supply and transport outlined above, what does the palaeo-dust record represent? Flux changes may record information on changes in the dust supply, reflecting changes at and/or of source area(s). Source areas may have expanded, through glacigenic action (Mahowald and Muhs, 2006), emergence of continental shelves into the sub-aerial zone (Bigler et al., 2006), and (seasonally-extended) expansion of arid and semi-arid areas (Werner et al., 2002; Tegen et al., 2002). Particle size changes may reflect changes in transport and/or source. Many palaeoclimate studies have used dust size as an indicator of wind strength in source regions (i.e. larger particles indicate stronger palaeo-winds, e.g. Rea 1995) or changes in the transport pathway (smaller particles indicate longer transport pathways, e.g. Lambert et al., 2008). Clearly, these arguments only hold if there is no change in the source size distributions, and no change in the source-sink distance. However, transporting winds may have altered in their trajectory and/or speed or gustiness (*see section 3.2.1*). Transport paths may have been effectively extended or shortened by changes in the degree of rain-out en route (Yung et al., 1996; Ruth et al., 2005; Lambert et al., 2008). Deposition rates on land may have varied in response to changes in vegetation and resultant trapping of dust by increased surface roughness (Marticorena and Bergametti, 1995; Marticorena et al., 1997).

Rates of dust deposition in sediment records are most often reported (e.g. Kohfeld and Harrison, 2001) as aeolian mass accumulation rates (MARs), calculated as:

734 MAR (g m^{-2} a^{-1}) = LSR x DBD x *f*

735

736 where LSR = the linear sedimentation rate (calculated as the thickness of the sediment
737 section (m)/ the time taken for the sediment to accumulate (yrs), DBD is the dry bulk
738 density of the sediment (g m^{-3}), and *f* is the fraction of the sediment that is of aeolian origin.
739 Figures 9a and 10a show aeolian MARs for the present day and the LGM, respectively, as
740 collated in our updated version ('DIRTMAP3') of the original 'Dust indicators and records of
741 terrestrial and marine palaeo-environments, DIRTMAP' database (Kohfeld and Harrison, 2001).
742 For comparison, figures 9b and 10b show modelled present day and LGM dust fluxes
743 (Mahowald et al., 2006). Here, we discuss LGM deposition rates (as archived in DIRTMAP3);
744 comparisons between these palaeo-data and the model simulations are discussed in section 6.2.

745

746

747 Figure 9.

748

749

750 Figure 10.

751

752

753 4.2. *The continental loess record*.

754 For the terrestrial environment, sequences of loess sediments provide key aeolian archives on
755 every continent. High-resolution loess/palaeosol sequences can provide new understanding of the
756 relation between aerosols and climate (e.g. Maher, 2007): they provide a record of dust
757 accumulation over large continental areas (Kukla, 1987; Thompson and Maher, 1995); they can
758 be dated directly (Roberts, 2008; Roberts and Duller, 2003), to obtain dust flux changes through
759 time; and they provide mineralogical, physical and chemical information on transported dust
760 (e.g. Prins et al., 2008; Sun et al., 2008), which in turn may affect biogeochemical cycles in the
761 ocean. Loess sequences can act both as a sink and a source for aeolian sediments. For example,
762 the Chinese Loess Plateau, the largest Quaternary accumulation of windblown sediment in the
763 world, has been identified by elemental and isotopic characterization as a major dust source for
764 the sedimentary record of the northwest and equatorial Pacific (Rea et al., 1994; Shigemetsu et

al., 2007; Ziegler et al., 2008). In turn, the glacial stage units of the Chinese Loess Plateau comprise extremely well-mixed dust sourced most likely by westerly transport from the western Taklimakan Desert and the northern margins of Tibet (e.g. Maher et al., 2009). The controversy over the extent of Tibetan glaciations during past glacial stages, and hence the extent of glaciogenic supply of loess, remains unresolved at present (e.g. Lehmkuhl and Owen, 2005).

Reflecting the proximal nature of many loess deposits to their sources, loess MARs at the LGM are the highest of any recorded aeolian fluxes (fig. 10a). For North America, for example, last glacial MARs in excess of 6,700 g m^{-2} a^{-1} have been recorded from loess sections in central Nebraska (Roberts et al., 2003). Provenance information indicates that these extremely high deposition rates reflect increased sediment supply resulting from desiccation and expansion of areas of the Great Plains, rather than from glacial production of silt-sized particles (Aleinikoff et al., 2008). In light of MARS typically ranging from ~ 100s to 1000s g m^{-2} a^{-1}, and notwithstanding their proximal and thus coarser-grained nature, loess transport on such a scale inevitably incurred deflation and entrainment of large volumes of finer particles (< 10 μm), of potentially significant radiative impact. For the central Nebraskan region, for example, PM_{10} MARs range from ~ 500 to > 900 g m^{-2} a^{-1}; such high rates of fine particle flux suggested to have caused prolonged post-LGM cooling in the central N. American region (Roberts et al., 2003). Elsewhere in N. America, loess was supplied from glacial sediment sources, downwind from the Rocky Mountains, and close to rivers issuing from the southern margins of the Late Wisconsin ice sheet. Significant loess accumulations span areas of Iowa, Illinois and Kansas; further north and west, parts of Idaho, Wyoming, Alaska and the Yukon peninsula (fig. 10a).

For the Eurasian region of the northern hemisphere, a swathe of loess girdles Eurasia from west to east (fig. 10a), reflecting dust supply both from expanded glacigenic and arid/semi-arid sources. For the European zone, loess MARs typically range from ~ 200 to 600 g m^{-2} a^{-1} for the last glacial stage, with some 'hotspots' of deposition reported, e.g. for the Nussloch site, Germany (> 2000 g m^{-2} a^{-1}, Rousseau et al., 2002) and Paks in Hungary (> 1000 g m^{-2} a^{-1}, Frechen et al., 1997). Extending eastwards over the Russian steppe, loess MARs have a similar range (~ 200 – 700 g m^{-2} a^{-1}), rates then increase towards Tajikistan (~ 1700 g m^{-2} a^{-1}, Frechen and Dodonov, 1998) and into the loess sequences of China. Across the Chinese Loess Plateau,

796 the increase in MARs during the last glacial stage, as estimated from median MARs across the
797 region, is of the order of up to x 5, i.e. ~ 300 g m^{-2} a^{-1} compared to an estimated 65 g m^{-2} a^{-1} for
798 the last interglacial stage (~ 125 ka, MIS5e) (Kohfeld and Harrison, 2003). However, the range
799 of LGM MARs estimated for the Loess Plateau spans < 100 to > 1000 g m^{-2} a^{-1}. In common with
800 most of the world's loess sequences, major spatial and temporal (sub-millennial) variability of
801 these glacial-stage increases in dust flux have been reported (e.g. Zhang et al., 2002). In terms of
802 mineralogy, quartz (~ 35 %), feldspars (~ 20 %), clay minerals (25 %, illite and illite-smectite)
803 and calcite (12 %) make up the very well-mixed loess (Jeong et al., 2008). Magnetic analyses
804 additionally quantify the presence of minor amounts of haematite and goethite (~ 1 %); the grain
805 size of the haematite in the interbedded palaeosols is distinctively sub-micrometre, and confers
806 the reddish hues to these soils (Maher, 2007).
807
808 Figure 11
809
810 A characteristic feature of many loesses is their trimodal particle size distribution, with
811 overlapping coarse and fine size modes (fig. 11, and e.g. Wang et al., 2006; Machalett et al.,
812 2008; Lim and Matsumoto, 2008). For the Chinese Loess Plateau, particle size end members
813 have been identified (Prins et al., 2007) comprising a 'sandy loess' component (modal size 63
814 μm), a 'silty loess' (modal size 37 μm), and a 'clayey loess' (modal size 22 μm). The two
815 coarser end members are thought to reflect saltation and suspension of 'local' coarse dust during
816 dust storms in spring; the finer end member inferred to represent higher level transport by
817 background and dust storm supply. For these sequences, increases in the proportion of coarse
818 particles has been interpreted as an increase in wind speed and thus of more intense winter
819 monsoon circulation (e.g. Xiao et al., 1995; Maher and Hu, 2006). The increased glacial-stage
820 MARs are also associated with increased particle size. At Luochuan, central Loess Plateau, for
821 example, the median diameter of aeolian quartz particles is ~ 15 μm for the last glacial stage
822 sediments compared with ~ 6 - 9 μm for the Holocene (fig. 12; Xiao et al., 1992). These paired
823 increases in fluxes and particle size may indicate both expanded source areas and increased wind
824 speeds, possibly coupled with a shortened transport path. Asian dust is and has been a major
825 component of the sedimentary record of the Pacific Ocean (*see section 4.3*). Downwind of the
826 Loess Plateau, present day estimates of dust flux range from ~ 6 to 12 × 10^6 tons of Asian dust

827 transported annually to the central North Pacific. Larger quantities are probably deposited over
828 the western North Pacific, closer to the Asian sources (Uematsu et al., 1983).
829
830 Figure 12
831
832 For the southern hemisphere, significant loess accumulations exist in South America and New
833 Zealand (fig. 10a). For the South American continent, the loess sequences of the Argentinian
834 pampas (the central Argentinian plains) cover ~1.0 x 10^6 km^2, with an average thickness of ~ 30-
835 40 m, thus comprising the largest loess accumulation in the southern hemisphere. Many of these
836 sequences have yet to be analysed in detail; age control is limited for the Tezanos Pinto
837 Formation, traditionally considered to represent the last glacial stage but possibly extending to ~
838 145 ka (Kemp et al., 2004). Estimated/adjusted MARs for the Pampas for the LGM are thus
839 scarce but range from 45-145 g m^{-2} a^{-1} (Krohling and Iriondo, 1999, cited in Mahowald et al.,
840 2006). Primary sources of these aeolian sediments are linked directly or indirectly to the Andean
841 region and the *arid diagonal* located to the west. In general, the composition of the Argentine
842 loess mirrors the mineralogical and chemical composition of Andean rocks (Gaiero, 2007).
843 Reflecting a dominant Patagonian dust source, the southern loess deposits (~ 36-38 °S) have
844 similar chemical and isotopic composition and a dominance of smectite, with illite present in
845 smaller amounts (Gaiero et al., 2004). This contrasts with the clay mineralogy of northern loess
846 deposits (~ 26-28 °S), dominated (Schellenberger and Veit, 2006) by illite (70–85 %), followed
847 by smectite (10–25 %). Between the northern and southern loesses, illite slightly dominates the
848 mineralogical composition of loess, while the coastal plains (Buenos Aires province) exhibit an
849 approximately equal mixture of smectite, illite and kaolinite (Zarate, 2003). The loess deposits
850 are coarse-textured. In the southern Pampas, the average sand–silt–clay content is 44 %, 26 %
851 and 24 %, respectively, the maximum sand content (mostly very fine sand) being ~ 80 % (Bidart,
852 1996). Magnetic properties of loess from this area indicate the presence of detrital magnetite and
853 titanomagnetite as the main ferromagnesian minerals (Orgeira et al., 2009). Northern loess
854 deposits show fairly uniform particle size (silt loam) averaging 14 % sand, 74 % silt, and 12
855 % clay. 80 % of the iron in the sediment is in paramagnetic minerals; ~20 % of the iron is in
856 hematite (~1.6 % of the sediment's mass), and << 1 % of the total iron is present as
857 magnetite/maghemite (~0.1 % of the sediment's total mass) (Carter-Stiglitz et al., 2006).

858 Elsewhere in the southern hemisphere, loess occurs most extensively in New Zealand, notably in
859 the lowlands surrounding formerly glaciated areas of the South Island (Eden and Hammond,
860 2003). Glacial and floodplain sources have dominated dust production in New Zealand; its loess
861 deposits are thicker (or possibly better documented) than in the S. American Pampean region, for
862 example. Locally, Holocene loess has accumulated up to several metres thickness and very thick
863 (> 10s m) LGM accumulations have been documented (Berger et al. 2002). MARs for the LGM
864 range between 70 to 150 g m^{-2} a^{-1} (Eden and Hammond, 2003). Most of the New Zealand loess is
865 quartz- and feldspar-rich. Glacially-derived loess deposits are strongly weathered (Marx et al.,
866 2005), being clay-poor but mica-rich and relatively coarse-grained compared to dust derived
867 from arid lands (McGowan et al., 2005).
868 Loessic sediments occur to more limited extents in other southern hemisphere countries. The
869 'parna' of southeastern Australia, for example, is characterised by low supply and deposition
870 rates; it dominantly comprises illite and kaolinite, carbonate and silt-sized quartz particles (e.g.
871 Summerell et al., 2000). Accumulation rates measured at a 3m loess section in the highlands
872 west of Sydney were relatively unchanged from 50-10 ka, at between 28 - 45 g m^{-2} a^{-1} (Hesse et
873 al., 2003).
874
875 *4.3. The palaeo-dust record of HNLC regions in the Pacific.*
876 As discussed above, the dust-generating regions of E. Asia appear to have expanded and
877 contracted across a range of glacial/periglacial and arid zone environments during the LGM. In
878 turn, these source changes have produced major changes in dust flux and dust characteristics to
879 the downwind continental sequences of the Chinese loess and palaeosols, and thence onwards to
880 the northwestern and equatorial Pacific.
881
882 For the northwest and the equatorial Pacific regions, there are globally important questions
883 regarding the possible causal role of changes in ocean biogeochemistry and export production
884 (carbon sequestration, i.e. biomass permanently removed from the carbon cycle) during the
885 glacial and interglacial stages of the Quaternary. The importance of understanding the influence
886 of aeolian-supplied Fe can be illustrated by recent fertilization experiments in the northwest
887 region of the Pacific, where iron-enrichment experiments led to the greatest expansion yet
888 observed of phytoplankton standing stocks, together with shifts to dominance of large centric

889 diatoms (Tsuda et al., 2003; de Baar et al., 2005). These two features suggest that the NW Pacific
890 Ocean may be particularly sensitive to iron fertilization. With regard to export production (i.e.
891 subsurface storage, rather than re-release from the ocean mixed layer), the drawdown of
892 atmospheric carbon resulting from this experiment is estimated at ~ 150-1500 C atoms per iron
893 atom added (*see section 3.1*; de Baar et al., 2008). The controversial question of whether changes
894 in palaeo-dust fluxes have caused past changes in ocean productivity in the NW Pacific is also
895 outlined here. Unequivocal identification of palaeo-productivity changes in the Pacific remains
896 challenging in light of: differing approaches to determining sediment fluxes; varying rates of
897 formation and dissolution of individual biogenic components (e.g. carbonate and opal; Honjo et
898 al., 1995); and varying interpretations of radionuclide (e.g. ^{230}Th) and organic degradation
899 products (e.g. biogenic barite). Combined, these issues have resulted in difficulties in quantifying
900 and integrating different biological products and hence total marine export production. As
901 discussed below, different proxies and different approaches to age models have resulted in
902 differing interpretations of glacial-interglacial changes in palaeo-productivity.
903
904 *4.3.1. Northwest Pacific dust fluxes*
905 Reported as one of the best records of aeolian flux from the NW Pacific, core V21-146 from the
906 Shatsky Rise (37.7ºN 163ºE) was retrieved from an elevated core site which lies beneath the
907 loess-transporting westerlies and provides a robust δ^{18}O record for age control (Hovan et al.,
908 1989, 1991). These Shatsky Rise sediments display increased dust fluxes (~ x 3.5, i.e. from ~ 2
909 to 7 g m^{-2} a^{-1}) during glacial stages, and indicate a direct link between intervals of accelerated
910 continental deposition of loess and enhanced dust flux to the open ocean. Variations in aeolian
911 grain size in the Shatsky Rise (fig. 13) show forcing at Milankovitch orbital frequencies, but
912 coarser grains (median diameter ~ 8 μm) show some association both with interglacial stages (at
913 the 100 ka frequency) and with low tilt and maximal precession, i.e. associated with glaciations
914 (Rea, 1994). Gravity core H3571 was recovered from the Hess Rise (34.9 ºN 179.7 ºE) some
915 1500 km to the east of the Shatsky Rise site and ~ 3000 km from the Chinese mainland. This
916 record also displays higher dust fluxes during glacial stages, of similar magnitude to the V21-146
917 record. However, these two records show significant differences in the timing and magnitude of
918 their dust flux maxima; V21-146 has maximal fluxes at the LGM, for example, while H3571
919 displays only moderate flux values (~ 5.5 g m^{-2} a^{-1}) at this time.

Tracing Asian dust around its entire northern hemisphere trajectory, the Chinese loess, Pacific deep-sea record and Greenland ice cores all display large and rapid changes both in dust flux and particle size from interglacial into glacial climate stages (fig. 12). It is notable too that large dust flux changes are also evident *within* glacial stages, on sub-millennial timescales. The relative significance of changes in source (e.g. stronger and/or more frequent dust emission events) compared with changes in transport efficiency (e.g. shorter transit times and/or reduced rain-out en route) has been the subject of debate (e.g. Nilson and Lehmkuhl, 2001; Ruth, 2005; Lambert et al., 2008; Fischer et al., 2007). For the North-Greenland Ice Core Project (NGRIP) core, at the distal end of this transport path, the dust source is thought from isotopic evidence to have remained 'constant' through glacial and interglacial stages, i.e. the Taklimakan and Tengger/Mu Us deserts of W. China and Inner Mongolia, respectively (Svensson et al., 2000; Bory et al., 2003), though this is poorly expressed by models (e.g. Andersen et al., 1998). Given stationary sources, the evidence for increased dust flux all along the transport pathway, i.e. from the proximal Chinese loess sequences (e.g. Xiao et al., 1999; Ding et al., 2002), and across the mid-latitude Pacific (e.g. Hovan et al., 1991; Kawahata et al., 2000) does indicate stronger and/or more frequent winds at source (Ruth, 2005). This conclusion requires careful examination, however. Care is needed in linking the more proximal loess sequences with the distal Pacific record. First, from air mass back-trajectory analysis, present day (coarse, ~ 2- 20 μm) dust supply to the Loess Plateau appears dominantly controlled by ambient (non-dust storm) northwesterly surface winter winds (Zhang et al., 1999). This is in contrast to the (fine-grained) LRT dust, which is uplifted during springtime dust storms from sources such as the western Taklimakan Desert (Bory et al., 2003) and carried by the westerly jet at upper levels (~ 5- 8 km) before descending towards the northeasterly trade winds, to the south and east of the subtropical high (Zhang et al., 1999). In contrast, during glacial stages, the dust source for both the LRT dust and the Loess Plateau appears, on multi-proxy elemental and magnetic grounds, to have been the same, dominantly the western desert and Tibetan Plateau regions (Maher et al., 2009; Torii et al., 2001). Second, in terms of particle size, glacial-interglacial variations of particle size along the proposed dust trajectory - from the East Asian deserts to the Loess Plateau to the NW Pacific to Greenland - present some complications. For the Loess Plateau and for NGRIP, glacial stages are associated with increased particle size (fig. 12). Glacial stages in the NGRIP dust record are characterized by particles > 1.6 μm in diameter; interglacials by particle sizes ~ 1μm.

For the NW Pacific (Shatsky Rise), in contrast, finer sizes (median diameter ~ 3 μm) have been reported for glacial stages and coarser sizes (5 μm) during interglacials (e.g. Hovan et al., 1991), whilst data collated by Zhang et al. (2007) show no clear glacial-interglacial trend (fig. 13).

Figure 13

4.3.2. Equatorial Pacific dust fluxes

Until recently, consistency of glacial-interglacial records of dust flux has been difficult to obtain for the equatorial Pacific, reflecting the confounding effects of sediment redistribution by bottom water currents (e.g. Mitchell and Lyle, 2005) and inputs of volcanically-derived aerosols (Olivarez and Owen, 1991). Critically, glacial to interglacial aeolian fluxes estimated from oxygen isotope-derived age models have varied both in sign and magnitude (e.g. Rea, 1994; Murray et al., 1995). Development of the ^{230}Th normalization method (Bacon, 1984; Francois et al., 1991; Francois et al., 2004) may provide a means (Lyle et al., 2007; Broecker, 2008) to calculate truly aeolian MARs. This method is based on the assumption that the rain-out rate of particulate ^{230}Th sinking to the sea bed is equivalent to the known rate of ^{230}Th production by ^{234}U decay in the overlying water column. MARs are calculated by dividing the production rate of ^{230}Th in the water column by the concentrations of scavenged ^{230}Th in the sediment. In tandem, terrigenous dust fluxes have been estimated for these sediments by using the ratio of ^{232}Th) and ^{230}Th as a proxy for dust flux. ^{232}Th is dominantly sourced from continental dust sources (its concentration in volcanogenic materials, for example, is an order of magnitude lower), it is almost insoluble in seawater, and is not subjected to any post-depositional redox-related change. Differences in dust provenance have so far been reported to exert relatively little effect on measured ^{232}Th concentrations, falling within 1 ppm of the upper continental crustal average of 10.7 ppm (Anderson et al., 2006). For an increasing number of sediment cores spanning the equatorial Pacific, recent studies making use of these techniques consistently demonstrate increased dust fluxes during glacial stages, with dust fluxes up to 2.5 x higher during the last glacial stage (Anderson et al., 2006; McGee et al., 2007, Winckler et al., 2008). Dust fluxes decrease from west to east, and towards the south, where northern and equatorial South American dust sources may contribute (Anderson et al., 2006), compared to the dominantly Asian-sourced aerosol towards the north (Ziegler et al., 2009). These new data

indicate a coherent response by dust generation and transport processes, from the equator to the South Pole, to glacial/interglacial climate changes through the late Pleistocene (fig. 14). The gradient in dust flux values from north to south across the equator may change through time in response to changes in the latititudinal position and/or the intensity of the intertropical convergence zone (Rea, 1994; Rea and Hovan, 1995; McGee et al., 2007; Ziegler et al., 2008). As noted by Anderson et al. (2006) and Winckler et al. (2008), the observed equatorial Pacific glacial flux increase (up to x 2.5 higher) is very different from modelled dust fluxes for this region. For example, while earlier models (Mahowald et al., 1999, Reader et al. 1999) produced unrealistically large LGM flux increases (LGM/modern ratios ranging between 12 and 105), more recent simulations result in disproportionately low LGM fluxes and also a west to east *increase* in flux (Mahowald et al., 2006).

Figure 14

4.4. The palaeo-dust record of the HNLC ocean regions: Antarctic ice cores and Southern Ocean.

4.4.1. Antarctic ice cores

Some of the longest, most detailed palaeo-dust records have been retrieved from the Antarctic polar region. Our interpretation of this paleo-record would be greatly facilitated by measurements of present-day dust transport. Unfortunately, there are remarkably few dust data in the Southern Hemisphere and, consequently, a major problem in quantifying LGM/modern dust flux ratios for the wider Southern Ocean region is the paucity of data with which to assess present day dust fluxes from any of the S. hemisphere land masses (*section 2.2*). Dust fluxes recorded in Antarctic ice cores show that across the southern polar region dust flux was ~ 30 x higher during the LGM (fig. 15). The reasons for this very large increase in dust at the LGM can be multiple. Source regions over Patagonia were certainly extended due to drier conditions and the decrease in sea-level that left exposed wide continental regions. In addition, the transport paths could have been changed and the wet deposition is likely to have been considerably reduced. These factors taken in combination or seldom can explain the differences seen in the deposition fluxes in the ice cores (Yung et al., 1996).

Figure 15

The growing number of ice cores retrieved from the Antarctic plateau provide long, undisturbed dust and climate records (extending back over eight glacial cycles) representing not only the Southern Ocean region but also sectoral differences (fig. 16) within that region (Fischer et al., 2007). High resolution dust flux records can be compared between several locations, including Vostok (78° 28´S 106° 48´E) (Petit et al., 1999) and the two EPICA sites (EPICA community members, 2006), Dome C (EDC, 75°06′S, 123°21E, in the Indian Ocean sector) and at Kohnen station in Dronning Maud Land (EDML: 75°00′S, 00°04′E), and Talos Dome (72° 48´S 159° 06´E; Delmonte et al., 2009). For all these cores, the dust profiles are extremely similar both in terms of total dust flux and the magnitude of the glacial/interglacial changes (Fischer et al., 2007; Lambert et al., 2008) with high dust fluxes during glacial stages; up to 30 x higher compared with the Holocene and earlier interglaciations. At EDC, total dust fluxes range from ~ 0.4 mg m^{-2} a^{-1} at the Holocene to 12 mg m^{-2} a^{-1} during glacial maxima. Tight coupling of the dust and climate records is especially evident during cold intervals of glacial stages (i.e. when $\delta D > \sim 425$ ‰, Lambert et al., 2008). Episodes of warming are linked with reduced dust fluxes; for instance, each of the Antarctic isotopic maxima (EPICA community members, 2006) is associated with a decline in dust fluxes at both the EDC and EDML sites (Fischer et al., 2007), indicating synchronous changes in the dust source regions (e.g. reduced dust supply and/or reduced wind speeds).

Figure 16.

Comparison of the EDML and EDC records identifies that the flux of non-sea salt calcium (i.e. as a mineral dust proxy) was 3 x higher at the EDML, Atlantic sector site; this higher flux is attributed to its greater proximity to S. American dust sources (Fischer et al., 2007). Debate is ongoing with regard to the relative influences of glacial-interglacial changes in source strength and changes in dust transport processes (especially atmospheric residence time and transport rate) on rates of dust deposition at Antarctica (e.g. see Fischer et al., 2007 compared with Lambert et al., 2008). If source strength has been the major influence, then glacial dust fluxes to

1044 the Atlantic sector of the Southern Ocean, lying between the S. American sources and the
1045 Antarctic region, should approach those observed in Antarctica (*see section 4.4.2*).
1046
1047 An additional if subtle complication arises with reported regional differences dust particle size
1048 around the different Antarctic sectors (Delmonte et al., 2004). In terms of dust particle size, the
1049 mass-size distribution of aeolian particles in east Antarctica is well sorted around a mean mass
1050 diameter of 2 μm, the largest particle diameter is ~ 5 μm, and the contribution of particles < 0.7
1051 μm contributes < ~10 % of the total mass (Delmonte et al, 2004b). However, during glacial
1052 stages, smaller particles were deposited at the EDC site, in contrast to Vostok and Dome B,
1053 where increased particle size has been reported during glacial stages (Petit et al, 1982; Delmonte
1054 et al. 2004).
1055
1056 Provenancing of the Antarctic dust by magnetic (Maher, 2009), elemental (Marino et al., 2008)
1057 and isotopic, Sr and Nd, techniques (e.g. Grousset et al., 1992; Basile et al., 1997) is still being
1058 refined, in order to extend the range of well-characterised potential source areas. As noted by
1059 Mahowald et al. (1999), no potential source area can be discounted or verified if it has not been
1060 sampled and characterised. It is also essential to take into account particle size effects on
1061 mineral, elemental and isotopic signatures (figure 17a and b and e.g. Delmonte et al., 2004;
1062 2009). For dust both at Vostok and EDC, the isotopic fields are almost identical (within the
1063 ranges $0.708 < {}^{87}Sr:{}^{86}Sr < 0.711$ and $-5 < \varepsilon Nd\ (0) < +5$), indicating a common dust source across
1064 the East Antarctic region. So far, around 60 potential source areas samples from S. America (33
1065 samples), New Zealand (15), Australia (22), southern Africa (6) and the Antarctic Dry Valleys
1066 (6) have been subjected to isotopic analysis of their fine size fractions (< 5 μm), encompassing
1067 loess and loess-like deposits, silts and sands, fluvioglacial sediments, soils, moraines (Delmonte
1068 et al., 2004, 2007, 2009) and glacial lacustrine deposits (Sugden et al., 2009). At present, those
1069 potential source areas showing partial overlap with the glacial stage ice core dusts are S.
1070 America, New Zealand, the Antarctic Dry Valleys (fig. 17), and some Australian areas (Lake
1071 Eyre). The overlap with Australian-sourced samples increases for interglacial stage samples
1072 (Revel-Rolland et al., 2006; Delmonte et al., 2007; 2009; Marino et al., 2008).
1073
1074 Figure 17.

So far, the consensus has been that Patagonia, and possibly the Puna Altiplano area, have been the major glacial dust sources for the Antarctic ice records (Gaiero, 2007; Delmonte et al., 2007). Satellite observations identify these hypothesized source regions as prominent present-day dust sources (*see fig. 3b, section 2.2*). Lowered glacial-stage sea level rendered Patagonia twice as large as today and glacial activity was extensive; this region was most likely an intense dust source during the LGM and previous glacial stages (e.g. Iriondo, 2000; Sugden et al., 2009). There is no published continental record of Quaternary dust accumulation from Patagonia, reflecting the high potential for wind erosion and poor geomorphic conditions for sediment accumulation. Rather, loess accumulation was concentrated further to the north and east in the pampas of Argentina (e.g. around ~ 30 °S, ~ 60 °W). Age control for these loess sequences has not been well established yet, but MARs at the LGM are estimated at between ~ 45 and 145 g m^{-2} a^{-1} (Krohling and Iriondo, 1999). Evidence for direct links between the Antarctic records of dust deposition and activating sources within the Patagonian region comes from chronological and isotopic data on glacial outwash sediments fringing the Straits of Magellan (Sugden et al., 2009). During the last glaciation, an Andean ice sheet developed between latitudes 44 °S and 55 °S. Outlet glaciers flowing eastwards from this ice sheet debouched into large outwash plains, with meltwater eventually draining into the S. Atlantic Ocean. When the outlet glaciers advanced to terminate directly at the outwash plains around the (then emergent) Magellan Straits, large amounts of glacigenic dust were supplied and available for uplift and transport by intensified westerly winds. Conversely, when the glaciers terminated in proglacial lakes, the dust supply was effectively 'switched off'. Isotopic analysis (Sr, Nd) of the finest fraction (<5 μm) of Magellan glacial lacustrine sediments indicates that they may have been one of the sources of the Antarctic dust during glacial stages (Sugden et al., 2009). Correlation between the dated sequence of Patagonian glacial activity and the Antarctic (Dome C) dust peaks is good, although anomalies do occur (e.g. the ice core dust peak at 30 ka has no glacial correlative in Patagonia; and the dated sediments are lacustrine, possibly indicative of periods of glacial retreat rather than advance). Rapid retreat of the glaciers can account for the observed rapid decreases in dust flux during deglaciation. Notably, the deglacial decline in Antarctic dust flux precedes the reduction in sea ice in the Southern Ocean, the warming of the Southern Ocean and the deglacial rise in sea level (Sugden et al., 2009; Fischer et al., 2007; Wolff et al., 2006). One problem with this

proposed glacial outwash dust source is the key and characteristic presence of leaf wax-derived n-alkanes in the intervening sedimentary record of the Southern Ocean (*see section 4.4.2*); the presence of vegetation might suggest a more low-lying source area (e.g. the emergent coastal shelf) rather than a glacial/periglacial source.

4.4.2. The Southern Ocean dust record

It has so far proved difficult to quantify the flux of dust across the Southern Ocean, and thence to Antarctica, during past glacial stages. Glacial stage sediments are characterised by increased lithogenic MARs in all three sectors of the Southern Ocean but these sedimentary records have diverse terrigenous contributions, including iceberg-rafted debris, current-transported sediment and aeolian dust (Diekmann, 2007). As with the Pacific sedimentary record, significant lateral transport of sediment by strong bottom water currents has affected the Southern Ocean records. There are presently very few integrative studies which have used a range of independent proxies to establish robust lithogenic and aeolian MARs, and identify glacial/interglacial changes in export production. Martínez-Garcia et al. (2009) present multiproxy-derived estimates of dust and iron supply, and marine productivity, over the last 1.1My from a deep-sea sediment record (PS2489-2, ODP Site 1090) located in the subantarctic South Atlantic (fig. 18). The initial age model for this composite sedimentary record was obtained by oxygen isotope analysis of benthic foraminifera (prior to tuning by correlation between the sediment sea surface temperature record and the EDC temperature record). Using ^{232}Th:^{230}Th as a proxy for dust flux (in combination with other terrigenous tracers, including long-chain n-alkanes derived from leaf waxes), these authors report dust fluxes varying from ~ 1 g m^{-2} a^{-1} (~ 50 mg m^{-2} a^{-1} of iron) for interglacial periods, to ~ 5-7 g m^{-2} a^{-1} (~250 mg m^{-2} a^{-1} of iron) for glacial stages (fig. 18). Assuming the dust source to be S. America, these estimates thus indicate up to 7 x increase in the source strength for this region. This magnitude of change contrasts with the x 20-25 increase in dust flux recorded by the Antarctic ice cores (Lambert et al., 2008). These data suggest that increased dust source strength, due to increased (possibly glacially-driven) dust supply and/or higher wind speeds, might be less significant for the recorded Antarctic dust fluxes than increased efficiency of long range transport, due to shorter transit times and/or less rain-out en route.

Figure 18

In terms of dust source, whilst very few dust flux data exist for either the present day or the LGM for this region, present day dust fluxes seem very low (*see section2.2)*. Hence, it is probable that sources additional to present-day southern S. American source areas were active during the last glacial period. Recent research does indicate that southern S. America was the most important LGM dust source for this region, as evidenced by the dominant overlap of isotopic compositions of sediments from the Pampean loess (Gaiero, 2007), the Southern Ocean (Walter et al., 2000) and the Antarctic dust (Delmonte et al., 2008). Modelling (Krinner and Genthon, 2003; Lunt & Valdes, 2002) and mineralogical studies (Gaiero, 2007; Delmonte et al., 2008) indicate that ~ 80 % of dust reaching the Antarctic Plateau during cold periods was derived from Patagonia. Less clear is the contribution of southern S. American materials to the South Atlantic Ocean. On isotopic grounds, glacial sediments from the north Scotia Sea seem dominated by a southernmost Patagonian source (Walter et al., 2000). However, compared with the potential southern S. American sources, sediments from the Atlantic sector of the Southern Ocean (~41 °S, 14 °W) display notably higher chlorite content (Diekmann, 2007). Total detrital MARs for this region are ~15 g m^{-2} a^{-1} for the last interglacial stage (MIS 5, ~ 125 ka) and ~120 g m^{-2} a^{-1} for the LGM (Walter et al., 2000), but the aeolian contribution to these figures is not known.

For the other S. hemisphere land masses, the Murray-Darling basin was a major Australian dust source in the LGM, exporting dust towards the south and southeast (Hesse, 1994; Hesse and McTainsh, 2003). With dust storm frequency ~ 50 % higher (McTainsh and Lynch, 1996), the LGM flux of Australian dust to the mid-Tasman Sea (160 ºE, 40 ºS) is estimated at ~ 3 x Holocene levels (Hesse, 1994). An additional LGM sediment source was the ancestral Lake Eyre basin (Dulhunty, 1982). Australian dust may have been a long-term contributor to the Southern Ocean and the southwest Pacific (Stancin et al., 2006, 2008) but the record is obscured by poor carbonate preservation in the deep Southern Ocean basins and reworking of sediments by strong Antarctic circum-polar circulation. In terms of particle size, LGM dust transported to the Tasman Sea from Australia (Hesse and McTainsh, 1999) comprises a minor coarse silt component (modal diameter 20-25 μm) and dominant fine silt and clay (< 4 μm). Compared to the present day, there is little change either in the mode or the size of the coarser particles, indicating little change in the strength of the zonal westerlies over Australia at the LGM. The palaeo-dust

contained within Tasman Sea sediments contains fine-grained haematite/goethite, the concentration of which varies directly with the dust concentration (Hesse, 1997). Thus, the flux of fine-grained haematite/goethite to the Southern Ocean and southwest Pacific and from Australia was greater in the LGM and previous glacials than in the Holocene and previous interglacials. (The volumetric flux of these minerals, however, has yet to be determined). Although New Zealand appears to be an insignificant source of dust to the Pacific sector of the Southern Ocean today (Prospero et al, 2002; fig. 3), it represents a (poorly documented) potential contributor of dust to the palaeo-Southern Ocean. Given its significant terrestrial accumulations of loess, New Zealand may have been a significant source of dust to the southwest Pacific and Southern Ocean at least during glacial intervals of the Late Quaternary. While the geochemical fingerprint of New Zealand dust overlaps glacial and interglacial Vostok dust signatures (figure 16; Delmonte et al., 2004, 2009), it is likely that New Zealand, like Australia, contributes more to the Pacific than other sectors of the Southern Ocean. It is notable that New Zealand's Taupo volcano has been excluded as a potential aerosol source for the Antarctic ice cores from Dome C (75°06´S 123°21´E) and Vostok (78°28´S 106°46´E), on the basis of its relatively high SiO_2 content (Narcisi et al., 2005). The absence of Taupo-sourced tephra from the last 200 ka of the Antarctic ice cores suggests that dust transport from the New Zealand/Australian region to Antarctica is inefficient (Delmonte et al., 2004; Narcisi et al., 2005). Yet Taupo volcanic dust has been found in Greenland ice cores (Zielinski et al., 1996); it seems unusual that there could have been no transmission of dust, at least to subantarctic Southern Ocean waters, given the extreme size of some eruptions. For example, in an eruption dated at ~ 26.5 ka, Taupo emitted ~1.4×10^{18} g of tephra (Mason et al., 2004), i.e. 1000 x the current global dust flux.

Given the current scarcity of present day or palaeo-dust flux and provenance data for the southern S. hemisphere region, model simulations remain to be validated. For the LGM, modelling of southern hemisphere dust deposition (e.g. Lunt and Valdes, 2002) suggests up to 15 x greater flux of South American dust in the Atlantic sector of the Southern Ocean (compared with up to 7 x flux as reported by Martinez-Garcia et al., 2009), and > 90 % contribution of S. American dust to deposition in the Atlantic and Indian Ocean sectors. In the Lunt & Valdes (2002) study, the modelled LGM flux of Australian dust exceeds the S. American value (presumably reflecting the greater area of the Australian continent) but the Australian dust is

dispersed over the entire Pacific sector of the Southern Ocean (Lunt and Valdes, 2002). Other modelled fluxes (Mahowald et al., 1999, 2006) indicate LGM/modern ratios in the Southern Ocean ranging from 0.5 to 2 (with dust sources as modelled for the present day) and 20 to 50 (with modelled expansion in dust sources at the LGM).

5. Palaeo-productivity changes during the LGM in the HNLC regions

5.1. Northwestern and equatorial Pacific.

In the context of iron fertilization, did the measured LGM enhancement of flux of Asian dust overcome iron limitation in the HNLC regions of the northwestern and equatorial Pacific, resulting in significant drawdown of atmospheric CO_2? Export production has previously been reported to have been globally higher during the LGM, with just one exception, the Southern Ocean (Kohfeld et al., 2005). Some studies have identified an association between glacial climate stages and increased productivity in the Pacific Ocean. For example, for core H3571 from the NW Pacific, biogenic opal and organic carbon were both reported to have increased during glacial stages (Kawahata et al., 2000). Similarly, for sediments retrieved from the eastern equatorial Pacific, Pedersen (1983) identified peak sedimentary organic carbon content (C_{org}) in association with the LGM. This study used the iodine:C_{org} ratio to rule out enhanced preservation as a causal factor and thus ascribed the peak C_{org} to a significant increase in productivity during the LGM. Using biogenic barite fluxes as a palaeo-productivity indicator and conventional stratigraphically-derived MARs, Paytan et al. (1996) also identified ~ 2 x higher export production during glacial stages for central and eastern equatorial Pacific cores.

More recent studies reporting increased glacial stage export production, in the western equatorial and subarctic Pacific, include those of Zhang et al. (2007) and Amo and Minagawa (2003), respectively. Zhang et al. (2007), using a mixture of micropalaeontological and foraminiferal isotopic ($\delta^{13}C$) proxies from Hole 807A (Ontong Java Plateau, 3°36.42N, 165°37.49E), find that productivity has been greater during glacial stages and has gradually increased since MIS13 (~ 530 ka). They also find correlation between their palaeoproductivity record and the dust flux record in northwestern Pacific core V21-146 (37ºN 163ºE), from the Shatsky Rise (Hovan et al., 1991). Zhang et al. (2007) thus suggest that dust transported from East Asia may be significant for enhancement of biological productivity in the western equatorial Pacific during glacial times.

Similarly, Amo and Minagawa (2003) estimated temporal changes of organic carbon fluxes during the last 130 ka from marine and terrigenous sources for sediments at the Shatsky Rise (33.3 ºN, 159 ºE). In contrast with the minor input of terrestrially-derived carbon, C_{marine} was estimated at > 86% of the total organic carbon for the whole core, reaching highest accumulation rates (9.2 mg C cm^{-2} ka^{-1}) in the LGM, and next highest rates in earlier glacial periods. From their alkenone-derived SST reconstructions, Amo and Minagawa (op. cit.) show that although cold sea surface temperatures (SSTs) prevailed at their Shatsky Rise site throughout MIS 2-4 (~ 20 – 7- ka), export production peaked at the LGM, when aeolian dust flux to the site was also at a maximum. It should be noted that both this study and that of Zhang et al. (2007) use conventional $\delta^{18}O$-derived chronologies for calculation of MARs.

However, recent work on equatorial Pacific sediments has contradicted these findings. Revising their previous conclusions, Paytan et al. (2004) reported that with normalisation of sediment accumulation rates using ^{230}Th, the apparent glacial increases in Pacific productivity either disappeared or even reversed. They conclude that higher rates of biogenic flux during glacial stages result not from greater export production but from increased lateral influx of sediment, transported by bottom water currents. This problem of sediment focusing appears to be widespread across the equatorial Pacific region, as indicated by ratios in excess of 1 for $^{230}Th_{measured}$:$^{230}Th_{expected}$ water column production). In another approach, using calcite accumulation rates (excess ^{230}Th-normalised and corrected for dissolution), Loubere et al. (2004) also estimated significantly reduced export production in the eastern equatorial Pacific (from 0.1 to 3.1 ºS and 84.65 – 95.65 ºW) at the LGM, i.e. ~ 30-50 % lower than during the Holocene. They attribute this reduction to lowered nutrient supply from the Southern Ocean via the equatorial undercurrent. This magnitude of productivity reduction is echoed by Bradtmiller et al.'s (2006) analysis of ^{230}Th normalized opal fluxes and ^{231}Pa:^{230}Th ratios in eleven equatorial Pacific cores; Holocene opal burial rates those of the late glacial period by 35 %. Rather than systematic reduction in productivity during glacial stages, Anderson et al. (2008) report that ^{230}Th-normalized barite fluxes (equatorial core, TT013-PC72, 140 ºW) exhibit no clear glacial–interglacial pattern of variability (fig. 19). Similarly, whilst acknowledging the absence of any 'ideal' palaeo-productivity proxy, Ziegler et al. (2008) identify no relationship between enhanced flux of Asian dust and ocean productivity, estimating similar export production rates at the LGM

as at present day (for five equatorial cores retrieved at 140 ºW). Nor do they find any relationship between palaeo-productivity and potential Fe supply by upwelling from the equatorial undercurrent.

For the subarctic, northwestern Pacific (ODP 882, 50.3 ºN, 167.5 ºE), Jaccard et al. (2005) report reduced export production during glacial stages (defined using a $\delta^{18}O$-derived age model), based on use of 'biogenic barium' as a palaeoproductivity proxy (i.e. sedimentary barite concentrations normalized by detritally-sourced aluminium). They attribute this glacial stage reduction in productivity to increased glacial ocean stratification, and reduced nutrient supply from upwelling. Their findings are supported by Shigemitsu et al. (2007) who report increased aeolian fluxes of loess during glacial stages but reduced marine productivity for another western subarctic Pacific core (50°N, 164.9°E) . Similarly, low glacial productivity (succeeded by increased deglacial productivity) is reported by Gebhardt et al. (2008), using biogenic opal and chlorins as proxy indicators for another piston core (51.27 ºN, 167.7 ºE) near to ODP Site 882.

At present, therefore, where sedimentation rates have been normalized by excess ^{230}Th, evidence suggests there has been no enhancement of ocean productivity and export production in the equatorial Pacific during the last glacial stage (fig.19). For the NW Pacific, varying with choice of proxy, contradictory evidence currently exists. As suggested by Gebhardt et al. (2008), productivity changes may have differed in time and space across the open ocean. However, selection and interpretation of productivity proxies in the Pacific region remain an ongoing issue and, as noted by Ziegler et al. (2008), it may be problematic to infer a change in total productivity from a change in any one particular biological component.

Figure 19.

5.2. Southern Ocean.

In terms of export production, Martínez-Garcia et al. (2009) report good correlation between glacial/interglacial changes in total organic carbon and the estimated fluxes of dust, indicating enhanced, and possibly dust-related, marine productivity in the subantarctic eastern Atlantic sector of the Southern Ocean during glacial stages (fig. 18). However, despite the high fluxes of

dust recorded by the Antarctic ice cores during the LGM, glacial increases in TOC appear much greater during previous glacial stages. For example, while the LGM is characterised by ~ 2 x increase in TOC, increases of up to ~ 28 x are observed during MIS 10 (~ 360 ka) and earlier glacial stages. One feature common to all the glacial intervals recorded in this composite record is that increases in export production (i.e. above the average interglacial value) only occurred when the ice core record of CO_2 concentration fell below ~ 230 ppmv (i.e. > 50 ppmv lower than during interglacial stages).

Figure 20

Enhanced export production in the glacial subantarctic has previously been reported, based on a range of radionuclide proxies, for each sector of the Southern Ocean, but varying spatially across the sectors (Kumar et al., 1995; Pondaven et al., 2000; Dezileau et al., 2003; Chase et al., 2001, 2003). At the present day, biogenic opal accumulates at similar rates in each sector, in the permanently open-ocean zone of the southern part of the Antarctic Circumpolar Current (ACC), with productivity maxima south of the modern Polar Front (fig. 20). During the Last Glacial Maximum, opal accumulation rates decreased in the southern part of the ACC at similar rates in all three sectors, and increased at regionally different rates in the northern part of the ACC, with maxima in the Atlantic sector (Diekmann, 2007). This suggests that limitation on primary productivity was overcome to a greater extent in the Atlantic than in the Indian and Pacific sectors (fig. 19). For the LGM, thorium-normalised lithogenic MARs were ~ 4 x higher in the Atlantic than in the Pacific sector (Chase et al., 2003). Glacigenic and bottom water-transported sediments are likely, however, to have contributed most of the fine-grained terrigenous material to this sector (Diekmann, 2007; Maher and Dennis, 2001; Latimer and Filipelli, 2001; Tagliabue et al., 2009).

Figure 20

6. Synthesis.

6.1 LGM dust fluxes and changes in ocean productivity?

1322 It is clear from the palaeo-data that dust fluxes to the HNLC regions of the world ocean were
1323 higher during the LGM than at the present day. The key question arises of whether or not these
1324 increased mineral fluxes resulted in significant changes in marine productivity, export production
1325 (i.e. carbon sequestration, biomass that is permanently removed from the carbon cycle) and,
1326 consequently, drawdown of atmospheric CO_2 during the last glacial stage.
1327
1328 As summarized above, a growing body of evidence suggests that there has been no enhancement
1329 of ocean productivity and export production in the equatorial Pacific during the LGM. For the
1330 NW Pacific, the evidence appears contradictory. The differences in data interpretation for this
1331 latter region may reflect heterogeneity in ocean productivity through time and space (Gebhardt et
1332 al., 2008), but choice and understanding of productivity proxy also remain an ongoing issue.
1333 Further, the relationships between productivity and export of carbon remain poorly resolved for
1334 this region. Data reported by Hays (2009), for example, indicate lower productivity at the LGM
1335 than in the Holocene but higher LGM export of carbon due to significant re-organization of the
1336 epipelagic food web, in particular the glacial dominance of more productive, deep-living (> 200
1337 m water depth) radiolarian species (especially *Cycladophora davisiana*), as previously reported for
1338 the Sea of Okhotsk (e.g. Hays and Morley, 2004).
1339
1340 For the Southern Ocean, coupling between dust flux and export production is inferred by
1341 Martinez-Garcia et al. (2009), who note that the observed increase in export production during
1342 glacial stages is non-linear, rising only slowly in early glaciation stages before accelerating to
1343 peak at glacial maxima. They attribute this exponential pattern to dust-driven iron supply, with
1344 dust flux also rising exponentially as global ice volume increases (Ridgwell and Watson, 2002;
1345 Lambert et al., 2008). These data would indicate that dust-forced increases in Southern Ocean
1346 export production can induce significant CO_2 drawdown but only during glacial maxima, and
1347 thus can account for ~ 40 -50 ppmv drawdown of CO_2 (i.e. as much as half of the total
1348 interglacial-glacial change of ~ 100 ppmv). The apparent coherence between the productivity
1349 proxy (alkenones) and the record of change in atmospheric CO_2 is notable (fig. 21c). Intervals
1350 where the relationship between dust flux, productivity and CO_2 break down (e.g. ~ 220 ka) seem
1351 quite rare but may provide some clue to the required additional causal factors in CO_2 drawdown.
1352 The coupling of dust flux and CO_2 in the reverse direction (i.e. glacial to interglacial) is less

clear. Rothlisberger et al. (2004) note that only small CO_2 variations accompanied large variations in the flux of non-sea salt-Ca^{2+} (as a proxy for dust) during glacial Antarctic warm events A1 to A4. They estimate that decreased dust deposition to the Southern Ocean during glacial to interglacial transitions accounts for not more than a 20 ppmv increase in CO_2 (with changes in the North Pacific adding <8 ppmv CO_2).

As yet, productivity proxies have yet to be quantified; there is no linear relationship between, for example, the Southern Ocean alkenones concentrations and palaeo-productivity. It is possible, however, to estimate some upper and lower bounds on the extent of productivity increase needed to account for the timing and fall in atmospheric CO_2, as recorded in the Antarctic ice cores (Box 1).

Similar calculations can be made for other waters. For example, if the drawdown of CO_2 was attributable to the HNLC waters of the NW Pacific as well as the Southern Ocean, then at ~15 % of the global NPP, the estimated glacial changes to NPP and/or pump efficiency would be proportionally smaller. Regardless of whether interglacial-glacial changes in atmospheric pCO_2 are due to a change in pump efficiency, a change in NPP or a combination of the two, the simplest case is for pump efficiency in the Southern Ocean to be towards the upper end of the probable range (i.e. closer to 20 % than to 1 %). Lack of observational data (as opposed to modelled estimates) prevents further resolution of the terms here and we suggest this is a high-priority goal for future research.

Figure 21.

However, whereas Martinez-Garcia et al. (2009) infer that dust-forced iron fertilization has been operating in the subantarctic region over at least the last 1.1 My, the spatial distribution of LGM increases in ocean productivity does clearly vary with sector. LGM opal accumulation rates increased in the northern part of the ACC but were greatest in the Atlantic sector of the Southern Ocean, coincident with highest detrital sediment accumulation rates. Rather than of aeolian origin, much of this terrigenous flux is likely to have been of glacigenic and/or bottom water-transported origin (Diekmann, 2007; Maher and Dennis, 2001). Further, even within this sector -

much of which lies directly downwind of the S. American dust source -, it is suggested that areas of enhanced LGM productivity existed as rather localized ‘hotspots’ (Anderson et al., 2008), and that these ‘hotspots’ may reflect localized iron inputs, such as reactive (i.e. more bioavailable), nanoparticulate iron from iceberg rafting (Raiswell et al., 2008).

BOX 1: Estimations of changes in net primary productivity and/or efficiency of the biological pump at glacial terminations.

1. Global net primary productivity (NPP) is ~ 105 Pg C per year, with ~50 Pg of this in the oceans (e.g. Field et al., 1998). The efficiency of the biological pump is contentious (e.g., Archer and Johnson, 2000; Moore and Doney 2007; Jin et al., 2008); estimates span a wide range from < 1 to 20 % of NPP being exported, depending both on the location and definition of 'exported'. Thus, modern carbon export may lie between *0.5 - 10 Pg C* y^{-1}.
2. From the onset of glaciations, it takes at least 10,000 years for CO_2 to drop by 100 ppm, such a decrease representing ~ 220 Pg carbon, or *~0.02 Pg C* y^{-1}.
3. Assuming that the decrease in atmospheric CO_2 was due solely to enhanced export from the Southern Ocean, bounding conditions can be established by considering two end member cases. The first end member case (see 4. below) is when NPP changes and pump efficiency is constant; the second end member case (see 5. below) is when NPP is constant and the pump efficiency changes.
4. Modern primary production in the Southern Ocean (waters south of 50 °S) is ~ *2 Pg C* y^{-1} (Arrigo et al., 2008). At the termination of a glacial stage, with a 1 % pump efficiency, total export = *0.04 Pg C* y^{-1}, i.e. the 'baseline' export of 1 % of the modern NPP (2 Pg) = 0.02 Pg C y^{-1}, plus the 0.02 Pg C y^{-1} 'extra export' required to reduce atmospheric CO_2 to its observed glacial value. This would require NPP at the glacial termination to have been 4 Pg C y^{-1}, i.e. a *100 % increase over the modern NPP value*.
5. Conversely, at a glacial termination, with 20 % pump efficiency, the total carbon export of (0.4 + 0.02) Pg C y^{-1} would have required an NPP of 2.1 Pg C y^{-1}; i.e. just *a 5% increase over modern NPP*.
6. In the case of constant NPP (at 2 Pg C y^{-1}), the total required glacial export in the Southern Ocean of 0.04 Pg C y^{-1} indicates a pump efficiency of 1.3 % (a 30 % increase over a baseline 1% efficiency), while total export of 1.22 Pg C y^{-1} gives a pump efficiency of 20.3 % (~2 % increase over a baseline 20 % efficiency).

In summary, changes in Southern Ocean marine productivity, during the later stages of the LGM, may have contributed up to half the recorded glacial fall in atmospheric CO_2. These changes may have been linked directly and causally with increased S. American dust fluxes, but are also likely to reflect iron supply from other detrital sources.

6.2 LGM dust fluxes and changes in radiative forcing?

Dust flux data retrieved from sediment archives (e.g. figs. 8, 10a, 14) demonstrate the major, global increase in 'dustiness' at the LGM, with at least some evidence of coherent MAR increases between both hemispheres, even at sub-millennial timescales. From the terrestrial loess sequences in particular, very high dust fluxes indicate marked increases in dust source 'strength', i.e. dust supply increasing due to significant expansion of source areas. Provenance studies of loess shows that this expansion reflects desiccation and expansion of arid and semi-arid areas (such as the American Great Plains, the Russian steppe), as well as continental- (northern hemisphere) and regional-scale (southern hemisphere) glaciation, acting to supply freshly ground rock particles for deflation and transport at and beyond the ice sheet margins (Hughes, 1992). Modelling studies also indicate that expansion of dust source areas is required to simulate the observed LGM fluxes. Using linked terrestrial biosphere, dust source, and atmospheric transport models to simulate the dust cycle, Mahowald et al. (1999), for example, show that the simulated increase in high-latitude dustiness requires the expansion of unvegetated areas, especially in the high latitudes and central Asia. This expansion results from increased aridity and lowered atmospheric CO_2, indicating the impact of vegetation feedbacks in modulating the atmospheric dust sources and fluxes. Visual comparison of the LGM modelled and measured dust fluxes (fig. 10), however, shows that while the dust cycle models simulate far-field dust deposition moderately well, they frequently under-estimate dust loadings close to dust sources, and over the continents in general, sometimes by up to an order of magnitude. Some of this discrepancy has been resolved by a twofold approach: firstly, by incorporating dust into a GCM and secondly, by including in dust cycle models a number of glaciogenic dust sources, designated by inversion from the palaeo-data (fig. 10b, and Mahowald et al., 2006). An important caveat is that dust cycle models do not commonly represent particles at the coarser end of the size distribution (> 10 μm). Present day observations of dust are often similarly constrained; many aerosol studies use cascade samplers with a cut-off at 2 μm. Thus, in making comparisons between model outputs

and dust observations and/or archives (e.g. sediment cores), it is important that size fractions are matched (e.g. Mahowald et al., 2006). Nevertheless, current dust cycle models have yet to adequately compute deposition fluxes in regions including or in the vicinity of dust sources (as above). The converse problem is that the models appear to over-estimate dust fluxes in the Southern Ocean, where dust deposition (to the Kerguelen Plateau) is up to 2 orders of magnitude lower than in most current models (Wagener et al., 2008).

Complementing these increases in supply of dust, the enhanced transport of that dust can readily be invoked, given – for example - that the Southern Ocean westerly winds are likely weaker and/or shifted equatorward during glacial periods compared to interglacial periods (Toggweiler et al., 2006). However, enhanced transport can be ascribed not only to more vigorous atmospheric circulation. Changes in transport path and removal mechanisms are also likely to have occurred, e.g. reduced rain-out (leading to increased atmospheric residence times), shortening of trajectory from an expanded source, and/or shifting of the atmospheric jet. Such transport factors may also have resulted in regional-scale variations in dust particle size, rendering subsequent inferences of wind speed changes subject to some degree of caution. Based on changes in dust particle size, LGM increases in wind speed have previously been inferred for northern and central China, Greenland, NW Africa, southern S. America and Antarctica. However, more complicated particle size/transport path relationships are evident, for example, for the dust record of the NW Pacific (despite its 'intermediate' position between the Chinese loess and the Greenland ice cores), and indeed across the Antarctic Plateau.

Intensified cyclonic activity in northern middle latitudes has been modelled for the LGM (e.g. Shin et al., 2003; Bush and Philander, 1999) although with some southerly displacement (~ 3°) of the core of the jet. Depending on the reconstructed ice sheet topography (Peltier, 1994), the Laurentide ice sheet also splits the jet stream in the above-cited models; such a split might have resulted in a more directly coupled transport path from the Chinese dust sources to the Greenland ice sheet.

In radiative terms, models have indicated rather different outcomes arising from increased glacial dust fluxes. For example, enhanced glacial fluxes of dust may have increased albedo resulting in

marked net cooling (Claquin et al. (2003); Mahowald et al. (2006)), in turn reducing convectional intensity and precipitation (Miller et al, 2004), and diminishing poleward transport of heat and moisture (Ivanochko et al., 2005). In direct contrast, Overpeck et al. (1996) suggested that high atmospheric dust loadings during glaciations may have resulted in episodic, regional-scale warming (of the order of ~5 °C), downwind of the major Asian and ice-margin dust source regions. The modelled degree of warming was greater at progressively higher latitudes, reaching peak values (+2.4 °C) for high-albedo (snow- and ice-covered) areas with high atmospheric dust loadings. Over such bright surfaces, the effects of dust absorption may dominate those of scattering, thus reducing short wave backscattering and enhancing long wave radiation (*see section 3.2.2*). The model simulations reported by Overpeck et al. (1996) also show glacial-stage warming patterns which differ markedly from the pattern of the applied, dust-induced radiative forcing. These differences likely reflect the interactive role of feedbacks, as atmospheric pressure, circulation and cloud patterns are altered, in modifying the regional energy balance. These authors note that these dust-driven radiative impacts may be under-estimated by the model, given the spatial and temporal variability of flux changes traced by the palaeo-dust record. Nor did the simulations include the radiative effects of deposited dust on snow and ice, which may significantly amplify warming effects (Peltier and Marshall, 1995; Krinner et al., 2006). Sub-millennial variability in dust fluxes may in turn reflect feedback-induced oscillations; for example, in the case of the Asian region, through modifying land-ocean thermal gradients, resultant monsoonal intensity and extent and type of vegetation cover in the semi-arid zone. Similarly, dust-induced warming in the Southern Ocean region may have caused glacial retreat in the southern S. America dust source areas, cutting off dust supply as glaciers terminated and deposited sediment in proglacial lakes rather than as unvegetated debris on outwash plains (Sugden et al., 2009).

Across a range of palaeo-dust records, it appears that dust fluxes peak just prior to Dansgaard/Oeschger oscillations, before and during Heinrich events, and just before glacial terminations (Rothlisberger et al., 2002). The Southern Ocean records suggest that up to half of the glacial-interglacial change in atmospheric CO_2 can be accounted for by dust-driven changes in marine productivity. It seems possible that radiative forcing by dust can provide an additional

1483 key feedback, producing warming above and beyond the extensive bright surfaces – whether ice
1484 and snow or bare soil and desert - of the glacial world.
1485
1486 *7. Conclusions.*
1487 Dust is important because not only can it affect climate, but the generation and transport of dust
1488 is itself extremely sensitive to climate (fig. 1, fig. 22). For the future, dust cycle models predict
1489 large changes in aeolian transport from the continents to the oceans over coming centuries, in
1490 response to anthropogenic climate change. It is thus important and timely to evaluate the extent
1491 to which changes in dust fluxes and properties may modulate or amplify global warming.
1492 Because spatial and temporal coverage of data on dust properties and emissions is limited at the
1493 present day, most of the currently reported dust budget values are based on transport models. The
1494 utility of these models and simulations is limited by uncertainties in our knowledge and
1495 understanding of dust sources and characteristics, and of atmospheric transport and dust removal
1496 and reaction processes (fig. 22). For example, regional-scale variations in dust composition,
1497 shape, particle size and surface roughness introduce significant uncertainties to the problem of
1498 modelling dust radiative forcing. At present, direct radiative forcing by dust appears to result in
1499 cooling over much of the Earth's surface; bright surfaces (such as deserts and ice and snow) are
1500 the exception, where dust can exert significant warming effects. *Improved modelling of dust*
1501 *optical properties at solar wavelengths can only be achieved with improved data on present and*
1502 *past regional variations in aerosol size and shape distributions, and in iron oxide*
1503 *concentrations, mineralogies and mixing state.*
1504
1505 Other types of dust cycle model are being developed in order to estimate the biogeochemical
1506 effects of dust transport to the oceans, through the supply of otherwise limited nutrients to
1507 marine phytoplankton. For example, an observational dust proxy (ocean surface concentrations
1508 of dissolved aluminium), a biogeochemical element cycling ocean model and a global dust
1509 entrainment and deposition model have recently been used in semi-independent combination to
1510 constrain dust deposition to the oceans (Han et al., 2008). However, even with major recent
1511 expansion of the ocean Al database, the present day spatial and temporal dust proxy coverage
1512 remains inadequate for resolving global dust climatology. This scarcity of data on dust fluxes
1513 and properties is an issue to be addressed with new field campaigns. In the case of ocean Al, new

sampling transects across the Pacific and Indian Oceans (within the CLIVAR and GEOTRACES programs), will begin to address one aspect of this problem. However, the most robust estimates of past and present dust fluxes are likely to be achieved only with use of multiple observational datasets (at optimized spatial resolution) to constrain dust cycle models (Cakmur et al., 2006). The DIRTMAP3 data (figs. 9a and 10a) clearly delineate those regions which remain severely under-represented in terms of any observed dust fluxes for the present day and/or the LGM. Such under-represented sites include much of the southern hemisphere, both terrestrial and marine. Even for those sites encompassed by DIRTMAP3, very few data exist regarding the key dust properties associated with the known sources and fluxes. *Enhanced collaboration between the modelling (radiative and biogeochemical) and data-gathering scientific communities can optimize design of new field campaigns to obtain key observational data both for the present day and for past time- (and climate-) slices*.

In order to achieve the required spatial and temporal information on dust fluxes, mineralogy, particle size and shape, effective and economically-feasible means of analysis are required. In terms of mineralogy, rapid elemental analysis using x ray-based techniques such as particle induced x-ray emission (PIXE), x ray fluorescence and automated scanning electron microscopy (with energy dispersive x ray analysis, e.g. 'QEMSCAN'), continue to evolve in terms of sample preparation, and lowering of analysis cost. For the iron-bearing components of dust, often nanoscale in size and present in minor or trace concentrations, non-destructive magnetic methods may provide both a sensitive and fast means of analysis. Regarding particle size, the first challenge for improving data on dust particle size distributions, whether at present day or for the palaeo-record, is the need for expanded geographic and temporal coverage. The second requires a degree of instrumental convergence between the atmospheric and oceanic communities. In particular it is notable that in the oceanic and ice-core community particle sizes are commonly measured with instruments based on the Coulter Principle (such as the Coulter Multisizer); such instruments are rarely used in aerosol research programs. Thus it is difficult or impossible to relate the size distributions measured by different research groups.

Not only is this a problem for those measuring dust fluxes but also in turn for the modelling community. If the cut-off size of the instruments used to measure dust fluxes is poorly defined,

then meaningful comparison subsequently with modelled dust fluxes is also hindered. *Thus, if one wants to say with confidence that a model is able to reproduce the main characteristics of the dust deposition fields at present and during the LGM period, then a strategy for measurement and treatment of dust particle size distributions is needed which is agreed and implemented across both the data-gathering and dust modelling communities.*

Similarly, just as convergence in analytical methods is at least desirable across the aerosol and palaeo-dust research communities, similar discussions between the aerosol, biological and palaeo-dust communities might aid development of the most appropriate analytical approaches to assessing the solubility and bioavailability of iron-bearing dusts.

The large changes in dust emissions and transport seen from the palaeo-dust record may reflect a variety of processes (fig. 22): changes in sources and source conditions; changes in vegetative cover; sub-aerial erosion of emergent continental shelves; deflation from periglacial deposits; variations in wind speed and gustiness; changed wind patterns linking sources to deposition areas; changes in deposition along the dust transport path. For the HNLC regions of the world ocean, the palaeo-dust record indicates increased dust fluxes at the LGM. The equatorial Pacific, for example, saw dust fluxes ~2.5 x higher during the last glacial stage, with dust fluxes decreasing from west to east, and towards the south. However, where sedimentation rates have been normalized by excess ^{230}Th, no enhancement of ocean productivity and export production has been observed in the equatorial Pacific during the LGM. The deep-sea record of the northwest Pacific, and the Greenland ice cores, display large and rapid changes both in dust flux and particle size from interglacial into glacial climate stages. It is notable too that large dust flux changes are also evident *within* glacial stages, on sub-millennial timescales. Contradictory evidence currently exists for productivity and export changes at the last glacial stage in the northwest Pacific region, interpretations vary depending on the type of proxy used and its interpretation. *More information is needed on the reorganization of marine ecology which results from glacial changes in ocean temperature and subsequent effects on export production*.

Dust flux data remain notably scarce in the southern hemisphere, both for the present day and for the past. *There is an urgent need for dust flux measurements in the Southern Oceans so that we*

can improve our understanding of dust processes in this region and to facilitate the development of atmosphere-ocean models. One practical approach to obtaining improved data coverage at the present day would be to install air samplers on the commercial and/or research vessels travelling between the Antipodes, South Africa, Antarctica and on islands in the Southern Ocean. For the past, the sedimentary record recently reported by Martinez-Garcia et al. (2009) appears to indicate up to 7 x higher dust flux at the LGM compared with the present day. This is in contrast to the much larger increase (x 30) recorded by the ice cores of the Antarctic Plateau. In terms of productivity, the Atlantic sector of the Southern Ocean appears to have responded most to these increased LGM dust fluxes, with increased export production having occurred slowly during the early glacial period before rising to a peak at the glacial maximum. This temporal pattern suggests that dust-forced increases in Southern Ocean export production can induce significant CO_2 drawdown but only during glacial maxima, and thus can account for ~ 40 -50 ppmv drawdown of CO_2 (i.e. as much as half of the total interglacial-glacial change of ~ 100 ppmv). Other drivers must be responsible for the early glacial decrease in atmospheric CO_2, i.e. from ~ 280 ppmv during the interglacial stage to the mid-glacial level of ~ 230 ppmv. It is possible to estimate some upper and lower bounds on the extent of the productivity increase in a) the Southern Ocean and b) the Southern Ocean and NW Pacific combined, in order to account for the timing and fall in atmospheric CO_2, as recorded in the Antarctic ice cores. These estimates suggest that the efficiency of the biological pump in HNLC waters is towards the upper end of the range (i.e. closer to 20 % than to 1 %).

Finally, it seems clear that enhanced collaboration between the diverse scientific communities - observational, modelling, biological – can only expedite resolution of the key gaps which exist in our understanding of dust sources, processes and climate impacts (fig. 22). Targeted and cross-calibrated collection of field data, both for the present day and past time- and climate-slices, seems a fundamental imperative. The aim must be to achieve not only robust measurement and/or estimation of dust fluxes across currently data-sparse regions, but also enhanced information on dust mineralogy, size, shape and surface roughness. Such data are essential both to benchmark model simulations and indeed to frame new dust, climate and biogeochemical models, including transient simulations and mesoscale modelling. *Until we have a good*

understanding of present-day processes, we will not be able to adequately address these processes either in the palaeo- record or with regard to the future impacts of dust on climate.

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Figure captions.

Figure 1. Schematic of the hypothetical glacial dust-atmospheric CO_2-climate feedback system. Different components of the Earth system can directly interact in three possible ways: positive influences (an increase in one component resulting directly in an increase in a second component) are indicated by red arrows; negative influences are indicated by black arrows; or no influence.. An even number (including zero) of negative influences within any given closed loop give rise to a positive feedback, operation of which tends to amplify any initial perturbation (e.g. the ice-albedo feedback). An odd number of negative influences gives rise to a negative feedback, which tends to dampen any perturbation. Primary interactions are indicated by solid lines, additional interactions dotted lines. Four main (positive) dust-atmospheric CO_2-climate feedback loops exist in this system. 1. Dust supply → ocean export production → xCO_2 → temperature → ice volume → sea level → dust supply (four negative interactions). 2. Dust supply → ocean export production → xCO_2 → temperature → hydrological cycle →vegetation → dust supply (two negative interactions). 3. Dust supply → ocean export production → xCO_2 → temperature → hydrological cycle → dust supply (two negative interactions). 4. Dust supply → ocean export production → xCO_2 → temperature → ice volume → dust supply (two negative interactions). From Ridgwell, 2002.

Figure 2. Dust deflation and entrainment processes (modified from Pye, 1987).

Figure 3. a) Remotely sensed aerosol optical depth measurements (Husar et al., 1997); b) The global distribution of major dust sources and dust transport paths based on the Total Ozone Mapping Satellite (TOMS) absorbing aerosol index (AAI) (Prospero et al., 2002). The figure is a composite of monthly mean TOMS AAI frequency of occurrence distributions computed using a threshold of 1.0 in the North Africa – Middle East – Asian dust belt and 0.7 everywhere else. The arrows show the main transport paths over the oceans. The arrow size is not indicative of the magnitude of the transport nor the distance that dust is carried. Some major points of note:

During the late boreal spring, summer and fall, huge quantities of African dust are carried to the western Atlantic and into the Caribbean and the eastern United States; also, from sources in the Arabian Peninsula and southwest Asia to the Arabian Sea and Indian Ocean. Dust is also carried across the Mediterranean to Europe and the Middle East.

Over the Atlantic during the boreal winter and early spring, dust from the Sahara and the Sahel is carried to South America; large quantities are also carried on occasion to the mid-latitude Atlantic and into Europe, as indicated by the north-looping arrow.

The large arrow in the North Pacific shows the main direction of large dust storms carried from Asia every boreal spring, in some cases penetrating to North America; the dust originates from a variety of sources including arid regions in Inner Mongolia (cross-hatched area) which do not show up well in the TOMS product.

The dust sources in the Southern Hemisphere are considerably smaller and less active than those in the Northern Hemisphere. The most active are those in Argentina, southern Africa and Australia.

Figure 4. a) Normalised size distributions of dust measured on Tenerife, Canary Islands (July, 1995) and in Puerto Rico (July, 2000) using the identical instrument, a TSI APS 3310. The Tenerife site is ~ 300 km from the N. African coast while Puerto Rico is 5000 km to the west.

The transit time for dust outbreaks is about 5 – 7 days; b) wind tunnel data (Alfaro et al., 1997) indicating smaller modal particle sizes entrained and transported at higher wind speeds.

Figure 5. Effect of the mass median diameter on dust deposition flux and on optical depth of the particle size distribution. Optical depth expresses the amount of radiation removed by scattering or absorption. Values are normalized to a size distribution with a mass median diameter of 4.5 μm and a sigma value of 2.1 (Reid et al., 2003, Table 1). A log-normal distribution with a constant width is assumed (Balkanski et al., 2007).

Figure 6. Transmission electron micrographs of discrete, pedogenic 'free iron' grains, a) typically acicular particles of goethite (αFeOOH), scale bar = 200 nm, b) more equidimensional particles of haematite (αFe_2O_3) - the smaller, electron-opaque particles -, essentially unassociated with the larger, clean flakes of kaolinite, scale bar = 1 μm; c) ultrafine haematite crystallite, soil sample, Morocco, scale bar = 100 nm. Micrographs from Maher et al. (2003), Jones (in Schwertmann, 1991), and Lieke (pers. comm.), respectively.

Figure 7. a) Atmospheric warming from absorption of radiation by dust; b) resultant net surface cooling. Integration of satellite and ground-based data for 2002 (from Chung et al., 2005).

Figure 8. Changes in glacial/interglacial dust concentrations over a pole-to-pole transect, spanning Greenland (the NGRIP ice core record, dust mass, ppm/10, Ruth, 2005, Svensson et al., 2000), the northern North Atlantic (Core T88, 48.3 °N, 25.0°W, high-field magnetic dust proxy, Maher, unpub. data), equatorial Atlantic (ODP Site 633A, 1.28 °N, 11.98 °W, terrigenous %, deMenocal et al., 1993) and Antarctica (EPICA Dome C, dust mass, EPICA Community members, 2004).

Figure 9. a) Measured aeolian MARs for the present day (for MAR data, see Appendix 1, and for MARs, age, site and quality control data, see the updated 'DIRTMAP3' database (Maher and Kohfeld, 2009, http://www.lec.lancs.ac.uk/research/LU_themes/inqua_working_group.php); b) modelled LGM MARs (Mahowald et al., 2006).

Figure 10. a) Measured aeolian MARs for the LGM (for MAR data, see Appendix 1, and for MARs, age, site and quality control data, see the updated 'DIRTMAP3' database (Maher and Kohfeld, 2009, http://www.lec.lancs.ac.uk/research/LU_themes/inqua_working_group.php); b) modelled LGM MARs (Mahowald et al., 2006 and Mahowald, pers. comm).

Figure 11. Typically trimodal particle size distribution of Chinese loess (Wang et al., 2007).

Figure 12. Time series of North Greenland Ice core Project (NGRIP) and Chinese loess data, synchronized on the NGRIP SS09 age scale (Ruth et al., 2003). Ice core data: $\delta^{18}O$, dust mass and mean particle size (diameter, lognormal mode of volume distribution); loess data: quartz mean diameter, from the central Loess Plateau site, Luochuan (Xiao et al., 1999).

Figure 13. Particle size records for the mid-latitude Pacific over several glacial-interglacial stages (redrawn from Hovan et al., 1991 and Zhang et al., 2007; 'v' in panel 3 = volcanic ash).

Figure 14. Time series of dust flux to the central equatorial Pacific (TTN013-PC72; 0.1°N, 139.4°W) and to the EPICA Dome C Antarctic ice core (from Winckler et al., 2008).

Figure 15. Dust fluxes at the Vostok and EPICA Dome C Antarctic ice cores (redrawn from Lambert et al., 2008).

Figure 16. The sectors of the Southern Ocean, together with the frontal system of the Antarctic Circumpolar Current, and the locations of the deep sea sediment and Antarctic ice cores referred to here (modified from Diekmann, 2007).

Figure 17. Isotopic characterization (Nd:Sr) of Antarctic dusts and suite of potential source areas, a) as reported for < 5μm potential source samples (Delmonte et al., 2004), and b) a refinement of plot a; progress definition of the Patagonian isotopic signature and inclusion of new samples from Australia (Revel-Rolland et al., 2006) and the Puna/Altiplano of South America (Gaiero, 2007; Delmonte et al., 2009) to explain the non-Patagonian isotopic signature found in the Antarctic dust.

Figure 18. Comparison of the Southern Ocean and Antarctic ice core records over the last 1.1Ma. (a) Temperature reconstruction from EPICA ice cores (black) (Jouzel et al., 2007) and alkenone-based SST from site PS2489-2/ODP1090 (red). Marine Isotope Stages (MIS) are shown for reference. (b) Atmospheric CO_2 concentrations from the EPICA ice cores. Dashed line indicates the CO_2 level when productivity starts to increase above the average interglacial value. Filled area illustrates CO_2 concentrations below 230 ppmv. Glacial terminations are shown for reference. (c) EPICA Fe flux (blue) and site PS2489-2/ODP1090 Fe flux (red). (d) EPICA Insoluble dust (light brown) and site PS2489-2/ODP1090 long-chain odd carbon–numbered n-alkanes (C25-35) mass accumulation rate (MAR) (blue). (e) Site PS2489-2/ODP1090 C37 alkenones MAR (green) and TOC MAR (Diekmann and Kuhn, 2002) (black). Shaded areas highlight the high-productivity intervals, when alkenone MARs are 3 x the average interglacial value. (Redrawn from Martinez-Garcia et al., 2009).

Figure 19. Time series of proxy records for palaeo-productivity and dust flux for the equatorial Pacific through several glacial-interglacial cycles (Redrawn from Anderson et al., 2008).

Figure 20. ^{230}Th-normalised fluxes of biogenic opal in the three sectors of the Southern Ocean (from Diekmann, 2007).

Figure 21. a) Global $\delta^{18}O$ stack(Lisiecki & Raymo, 2005) versus n-alkanes concentrations from Southern Ocean site PS2489-2/ODP1090; marine isotope stages shown; b) Temperature reconstruction from EPICA ice cores (Jouzel et al., 2007) and alkenone-based SSTs from PS2489-2/ODP1090; c) atmospheric CO_2 concentration from the EPICA ice cores (Luthi et al., 2008; Petit et al., 1999; Siegenthaler et al., 2005) versus alkenones concentration, PS2489-2/ODP1090.

Figure 22. Schematic of the sources, process and impacts of dust on climate, a) at the present day, and b) at the LGM, with key gaps in data and understanding marked by '**?**' on the diagrams.

Table captions.

Table 1. Comparison of a range of modelled present day dust fluxes (mean annual, by region). The unit is Tg/yr, numbers in brackets = the % of the annual mean global emission flux. (From Tanaka & Chiba, 2006).