Dust in the Earth system: The biogeochemical linking of land, air, and sea

ANDY J. RIDGWELL¹

School of Environmental Sciences, University of East Anglia, Norwich NR4 7TJ, UK Tyndall Centre for Climate Change Research, University of East Anglia, Norwich NR4 7TJ, UK ¹ Now at Department of Earth Sciences, University of California, Riverside, CA 92521, USA (A.Ridgwell@uea.ac.uk) Understanding the response of the Earth's climate system to anthropogenic perturbation has been a pressing priority for society since the late 1980s. However, recent years have seen a major paradigm shift in how such understanding can be reached. Climate change demands analysis within an integrated "Earth system" framework, taken to encompass the suite of interacting physical, chemical, biological, and human processes that, in transporting and transforming materials and energy jointly determine the conditions for life on the whole planet. This is a highly complex system, characterized by multiple non-linear responses and thresholds, with linkages often between apparently disparate components. The interconnected nature of the Earth system is wonderfully illustrated by the diverse roles played by atmospheric transport of mineral 'dust', particularly in its capacity as a key pathway for the delivery of nutrients essential to plant growth, not only on land, but perhaps more importantly, in the ocean. Dust therefore biogeochemically links land, air, and sea.

This paper reviews the biogeochemical role of mineral dust in the Earth system and its interaction with climate, and in particular, the potential importance of both past and possible future changes in aeolian delivery of the micro-nutrient iron to the ocean. For instance, should in the future there be widespread stabilization of soils for the purpose of carbon sequestration on land, a reduction in aeolian iron supply to the open ocean would occur. The resulting weakening of the oceanic carbon sink could potentially offset much of the carbon sequestered on land. In contrast, during glacial times, enhanced dust supply to the ocean could have 'fertilized' the biota and driven atmospheric CO₂ lower. Dust might even play an active role in driving climatic change; since changes in dust supply may affect climate, and changes in climate in turn influence dust, a 'feedback loop' is formed. Possible feedback mechanisms are identified; recognition of whose operation could be crucial to our understanding of major climatic transitions over the past few million years.

Keywords: dust; iron; climate change; carbon cycle; Earth system

1. Introduction

Solid particles of sufficiently small size (< 50 μ m) can be picked up from the land surface by the wind and may be carried great distances through the atmosphere. Although individual particles are often invisible to the naked eye, billions of tons of material are transported every year in this way. Transport events can often be of sufficient intensity to be visible from space, as shown in the accompanying satellite image (Figure 1). This 'dust' mostly comprises fragments of rock minerals and other soil constituents, but can include anything of suitable size, such as viruses and pollen grains. Industrial emissions also make an important contribution, particularly in the midlatitude Northern Hemisphere. Over the ocean, sea salt particles produced by wave action are a major constituent of atmospheric aerosol, although net deposition of this material tends to be restricted to coastal areas. For the purpose of this review 'dust' will be taken to be mineral aerosol, with a typical elemental composition given in Table 1.

Dust is entrained into the air from the land surface when the wind speed is sufficient to overcome the cohesive forces that exist between soil particles. Entrainment is facilitated by low soil moisture levels, when cohesion is minimal, and also by the absence of vegetation cover, which allows greater wind speeds to be reached at ground level. It is not surprising then to discover that dust sources are predominantly restricted to arid regions (particularly in association with topological lows) [*Prospero et al.*, in press]. While relatively heavy particles will tend to rapidly settle out of the air and be deposited close to their source, finer particles remain suspended in the air stream and are transported with the prevailing winds. Eventual deposition to the Earth's surface occurs through 'dry' depositional processes such as gravitational sedimentation or turbulent transfer, or 'wet', by entrainment into falling raindrops ('precipitation scavenging'). The influence of these factors in conjunction with atmospheric circularly high rates of deposition are observed immediately downwind from the Sahara and Sahel desert regions of

North Africa, extending out across the Atlantic to the Caribbean and northeastern South America, and also in the western Pacific and northeast Indian Oceans, associated with the deserts of central Asia. Less important dust sources located in Australia, southern Africa and Patagonia appear to have a more localized influence. In locations remote from any major sources of dust (such as the Southern Ocean and equatorial and south Pacific) in contrast, very low rates of dust deposition are found.

The presence of dust in the atmosphere affects its optical properties, reddening the apparent color of the sun and sky at sunset as the shorter, blue, wavelengths are preferentially scattered. By modifying incoming (ultraviolet and visible) and outgoing (infrared) radiation, the energy balance at the Earth's surface can be perturbed sufficiently to produce a local seasonal heating (over light-coloured surfaces) or cooling (over dark-coloured surfaces) of up to $\pm 2^{\circ}$ C [*Miller and Tegen*, 1998]. When deposited on snow cover, dust darkens the surface and decreases the fraction of sunlight that is reflected, producing an additional local heating effect. Another role of mineral aerosol is in its capacity to act as a carrier substrate. An apparent correlation has been noted between episodes of unusually intense dust deposition and severe coral decline in the Caribbean over the past few decades, with the suggestion being made that fungal spores associated with deposited North African dust disrupts the functioning of reef ecosystems [*Shinn et al.*, 2000]. In this way, dust may be facilitating the transport of pathogens across the ocean. Finally, dust in the atmosphere may have an important effect in influencing cloud nucleation. It is in modifying the flow of carbon and nutrients within the Earth system ('biogeochemical cycling'), however, that dust arguably plays its most fascinating and intricate role.

2. Dust deposition in the terrestrial realm

Wind-blown dust that settles on the land surface can accumulate to great thickness. For instance, over the course of the past few million years (Ma), dust carried east from the Gobi desert and deposited to the Loess Plateau region of China has resulted in the formation of soil sequences of

up to 200 m thick. Even in locations where deposition rates are considerably lower, soil structure can be affected. Where the underlying substrate is highly susceptible to weathering (e.g., the basaltic bedrock of the Hawaiian Islands), aeolian quartz often forms a major soil constituent. Since mineralogy and grain size in turn strongly influence the water- and nutrient-holding properties of a soil, dust can exert an important control upon ecosystem structure and plant productivity.

Dust can also play a much more direct biogeochemical role in terrestrial ecosystems. Tropical rain forests are extremely efficient ecological systems by virtue of the high degree of nutrient recycling and retention that occurs within the system. Despite low loss rates of important plant nutrients, in parts of Amazonia, the negligible input from already highly weathered (and thus nutrient-depleted) soils and limited riverine supply do not appear to be sufficient to maintain the nutrient balance on timescales of hundreds to thousands of years. This suggests that aeolian deposition of nutrients, particularly phosphorous (P), may be critical [Swap et al., 1992]. Dust transported across the Atlantic from the Sahara and Sahel deserts (such as occurs during periodic dust storms – see Figure 1) might then influence the maximum size of the ecosystem that can be supported. The highly weathered soils and phosphorous-limited ecosystems of some of the older (> 1 Ma) Hawaiian Islands suggest an analogous situation, with losses due to leaching and immobilization of this vital nutrient exceeding local supply [Kennedy et al., 1998]. Again, aeolian P transported across the open ocean (this time the Pacific) from remote sources (central Asia) is required to balance the nutrient budget of the system, and may thus provide a controlling influence on terrestrial productivity. Dust therefore directly links the land surface and ecosystems of two otherwise effectively unconnected landmasses on Earth.

3. Dust deposition in the marine realm

Mineral fragments are relatively insoluble in surface ocean waters, and settle largely unaltered through the water column to be deposited in sediments on the sea floor. As on land, accumulation

of this material can influence biogeochemical cycles. In places where dust fluxes are sufficiently high, the residence time of biogenic material such as the calcium carbonate and organic carbon in surface sediments (where it may be susceptible to degradation) is reduced. The presence of dust in deep-sea sediments can also subtly alter pore water chemistry, suppressing the susceptibility of biogenic opal to dissolution. By aiding preservation and thus enhancing sedimentary burial rates, dust deposition can accelerate the removal of various chemical species (including nutrients and carbon) from the ocean, with consequences for ocean productivity and the global carbon cycle.

Once again, there is a more direct and powerful effect of dust deposition. The major ('macro-') nutrients required by the primary producers of the open ocean (microscopic marine plants -'phytoplankton') are phosphate (PO_4^{3-}), nitrate (NO_3^{-}), and, for some species at least, silicic acid (dissolved silica; H₄SiO₄). As phytoplankton cells grow and divide in the sunlit surface layer of the ocean (the 'euphotic zone'), nutrients are removed from solution and transformed into cellular constituents. Most of this material is ultimately broken down ('remineralized') by the action of bacteria and zooplankton within the euphotic zone, returning the nutrients into solution. A fraction (in the form of dead cells, zooplankton fecal pellets, and other particulate organic debris) escapes and settles through the water column under the influence of gravity, being remineralized much deeper in the ocean. Although nutrients are eventually returned to the euphotic zone by upwelling and mixing, a vertical gradient is created with lower nutrient concentrations at the surface than at depth. This removal by the biota of dissolved constituents at the surface and export (in particulate form) to depth is known as the 'biological pump' (Figure 3). An important consequence of the supply of nutrients to the surface by the upwelling and mixing of nutrientenriched waters below is that delivery by dust does not appear to be a particularly important source of phosphate to the biota of the open ocean, in contrast to the situation that can occur on land. Similarly, the supply of aeolian nitrate and silicic acid by wind deposition is also typically of only minor importance. However, as we shall see, in some areas this is not the case for the micro-nutrient iron (Fe), with aeolian deposition playing a fundamental controlling role in the ocean carbon cycle and with it, perhaps, in the operation of the climate system.

(a) Iron limitation in the open ocean

A long-standing puzzle in oceanography has been why phytoplankton do not always fully utilize the macro-nutrients that are supplied to them. As shown in Figure 4, in certain areas of the world ocean (and in particular, the Southern Ocean), high concentrations of NO_3^- remain in surface waters. The situation is similar in many respects for both PO_4^{3-} and H_4SiO_4 (not shown). Despite the availability of NO_3^- , standing stocks of phytoplankton are relatively low, leading to the designation of such regions as 'High-Nitrate Low-Chlorophyll' (HNLC). Physical conditions (such as light levels) and the intensity of grazing by microscopic marine animals ('zooplankton') are likely to be at least partly responsible for the HNLC condition. Ever since the early part of the last century, it has also been strongly suspected that growth limitation through insufficient availability of iron might be critical. New results in the 1980s and early 1990s led to a vigorous debate (driven principally by the late John Martin) as to which of these factors provided the more fundamental control on phytoplankton growth.

Laboratory experiments conducted then demonstrated that the addition of Fe to HNLC water samples almost invariably stimulated phytoplankton growth and with it, increased NO₃⁻ uptake. Because the *in vitro* environment differs in a number of crucial respects from that of the ocean, the results of these small-scale experiments did not settle the debate. A methodology for carrying out Fe fertilization of the open ocean was therefore devised [*Watson et al.*, 1991], involving the dispersal of dissolved Fe from a ship whilst simultaneously marking the resulting 'patch' of enhanced Fe with an easily measurable label (typically the inert tracer sulphur hexafloride, SF₆). The use of a tracer is critical, since measuring ambient (sub nano-molar) concentrations of dissolved Fe in real time is extremely problematic, and the Fe-enriched patch can move 100s of km during the course of the experiment (typically 1-2 weeks). Following Fe release, the patch is crisscrossed, and observations made both within and outside the patch (as defined by the presence or absence of SF₆ in the water, respectively). The water outside acts as a 'control' on any changes measured in the Fe-enriched patch. One such experiment was carried out in February of 1999 in the Southern Ocean – the 'Southern Ocean Iron RElease Experiment' (SOIREE) [*Boyd et al.*,

7

2000]. As hypothesized, the phytoplankton responded to the addition of Fe with a strong increase in the concentration of chlorophyll a (a phytoplankton photosynthetic pigment, whose concentration can be taken as a rough indicator of cell density) within the patch, but not outside it (Figure 5).

In SOIREE, the impact of iron fertilization was so striking that the results of the experiment were visible from space! Six weeks after the initial Fe release, gaps in the cloud cover allowed the remote sensing of surface ocean optical properties. The processed ocean color satellite image (Figure 6) shows a clear 'bloom' of enhanced chlorophyll concentrations compared to the surrounding waters.

The theoretical potential that exists for removing CO_2 from the atmosphere ('sequestration') by stimulating phytoplankton growth through Fe fertilization has not gone un-noticed, and in anticipation of the possibility of 'carbon credit' trading in the future, a variety of deliberate Fe fertilization techniques have already been patented. However, the ultimate fate of the carbon incorporated into the surface biomass during transient artificial fertilization experiments such as SOIREE is not known, rendering the effectiveness of carbon sequestration by this method highly uncertain at present [*Ridgwell*, 2000].

(b) Iron supply to the surface ocean

Why should there be a deficit (relative to other nutrients) in the supply of iron to the biota, in some locations in the ocean but not others? Transport by rivers is the dominant route by which Fe is supplied to the ocean as a whole. Before it can reach the open ocean, though, rapid biological uptake and sedimentation in highly productive estuaries and coastal zones tends to remove much of the newly supplied Fe from the water. Rivers are therefore not thought to be an important source of Fe to the open ocean. As with the macro-nutrients, supply of Fe to the euphotic zone occurs through upwelling and mixing of ocean waters from below, which are enriched as a result of the remineralization of biogenic material (Figure 3). However, because the dissolved state of Fe is not thermodynamically favored in the oxygenated seawater environment, it tends to be

8

scavenged out of solution by attaching to particulate matter setting through the water column. The upshot of this is that there tends to be insufficient Fe in upwelled water compared to other nutrients such as the highly soluble NO_3^- . Aeolian deposition must supply this shortfall in order for NO_3^- to become completely depleted at the surface.

Inspecting the distribution of dust deposition to the ocean (Figure 2), it is clear that the fluxes to the Southern Ocean are amongst the lowest anywhere on Earth, primarily due to the relative lack of (ice-free) land area available in the Southern Hemisphere for sourcing dust. Aeolian supply is then not able to make up the shortfall (relative to NO₃⁻) in upwelled Fe. Here, then, lies the primary reason for the HNLC condition of the Southern Ocean. Similar reasoning also applies to the other major HNLC regions in the equatorial and northern Pacific Ocean.

(c) Iron supply and the global carbon cycle

Alongside factors such as ambient temperature, *p*H, and wind speed, the concentration of dissolved inorganic carbon (DIC) in the surface ocean exerts a fundamental control on air-sea CO_2 exchange. Processes that affect DIC concentrations will therefore influence the mole fraction of CO_2 (*x*CO₂) in the atmosphere, and with it, climate (via the 'natural greenhouse effect' – e.g., see *Houghton et al.* [2001]). One process that affects DIC concentrations is the biological pump. This is because along with nutrients, carbon is also taken up by phytoplankton in the euphotic zone and incorporated into cellular organic constituents, with a fraction later remineralized at depth (Figure 3). By reducing surface water DIC concentrations the equilibrium concentration of gaseous CO_2 is depressed, driving a net transfer of CO_2 from the atmosphere into solution in the ocean. The value of atmospheric *x*CO₂ will then exhibit an inverse relationship to the strength of the biological pump. Indeed, in the absence of any biological activity in the ocean, atmospheric *x*CO₂ would probably be more than 50% higher than it is today.

Variability (both in space and time) in aeolian iron supply already occurs naturally in the present-day Earth system – can a controlling influence of this on the biological pump be discerned? In a recent study of regional atmospheric circulation, the trajectories of air masses

originating in southern Africa and laden with iron-rich aerosols were analyzed, and it was found that subsidence of this air occurred in discrete patches in the central South Indian Ocean [*Piketh et al.*, 2000]. The locations of subsidence corresponded to hitherto unexplained regions of enhanced CO_2 uptake by the ocean (Figure 7), raising the possibility of a localized Fefertilization effect. On a wider scale, the 1991 Mount Pinatubo volcanic eruption resulted in the injection of a substantial mass of iron-rich material into the atmosphere. Subsequent measurements of tiny changes in the mixing ration of oxygen in the atmospheric are consistent with stimulation of biological productivity by this perturbation [*Watson*, 1997]. Evidence is therefore accumulating that natural variability in aeolian iron deposition affects productivity in the ocean and thus influences the pattern and magnitude of air-sea CO_2 exchange. However, since the residence time of iron in the ocean is of the order of several hundred years [*Ridgwell et al.*, 2002], the influence of such decadal (and shorter) variability in dust supply on the longer-term operation of the global Fe cycle and on the trend in atmospheric CO_2 will be comparatively restricted. We will now explore two examples of a rather more substantial and long-term dust perturbation in the Earth system and the potential climatic implications of them.

4. Anthropogenic modification of dust supply

As we have already seen, present-day supply of Fe to the biota of the HNLC regions of the ocean have been identified as being insufficient for the biological pump to work at its maximum efficiency (i.e., complete NO_3^- utilization). Furthermore, a number of other regions (such as of the central tropical Pacific and north Atlantic) are now suspected as being close to limitation or quasi-limited. Any reduction in dust supply can therefore be expected to intensify limitation where it already exists, and possibly produce a limitation of productivity where it was previous unrestricted. The result of this would be a reduction in the rate of CO_2 uptake by the ocean. This has clear implications for future atmospheric xCO_2 levels and thus for the degree and rate of future climate change. Under what circumstances might a reduction in dust supply to the ocean occur? One possibility is that should the changing climate result in a substantial reduction in the area of desert and semi-desert vegetation in the future, a weakening of dust supply to the atmosphere might be expected [*Harrison et al.*, 2001]. Working against this, population pressures are likely to drive an increase in soil disturbance via the intensification and extensification of agriculture. In addition to source changes, the efficiency with which dust is transported through the atmosphere may also change – increased precipitation scavenging of dust particles occurring under a more intense future hydrological cycle would result in a reduction in supply rates to the open ocean. However, we will consider a novel agent here, and in doing so, highlight a potential flaw in the carbon budgeting system proposed under the Kyoto Protocol.

The deliberate large-scale modification of terrestrial ecosystems has been identified as having considerable potential in the mitigation of climate change [*Royal Society*, 2001]. A variety of "land-use, land-use change and forestry" (LULUCF) activities have been proposed for the sequestration of carbon on land. These include, changes in soil management practices (for example, reducing tillage, enhancing the areal and seasonal extent of ground cover, and the 'set-aside' of surplus agricultural land), restoration of previously degraded lands, and forestation [*Royal Society*, 2001]. As a result of reduced disturbance and increased stabilization of soils, many of these activities are likely to lead to a reduction in dust supply to the atmosphere. Since dust exerts an important control on the biological pump in the ocean, the effectiveness of carbon removal from the atmosphere via sequestration on land may be diminished by a reduction in the quantity of carbon taken up by the ocean.

The potential importance of this previously unrecognized teleconnection within the Earth system, with deliberate actions taken on land producing unexpected side effects in the ocean, has been investigated with the aid of a numerical model of the ocean-atmosphere carbon cycle [*Ridgwell et al.*, in press]. The model is run with a prescribed time history of atmospheric xCO_2 (Figure 8a); observational values up until 1990, and following one possible future scenario (in which CO_2 in the atmosphere is stabilized at 550 ppm by 2150) thereafter [*Houghton et al.*,

2001]. As atmospheric xCO_2 changes, the rate at which the ocean takes up anthropogenic CO_2 is re-calculated (Figure 8b). The difference between control (dust fluxes to the ocean held at present-day rates) and perturbation (modified dust supply) runs then gives a measure of the impact of the perturbation on the global carbon cycle. Scenarios of a global reduction in dust flux to the ocean of 15% and 30% are tested, with the reduction ramped up over a period of 50 years starting in 2000.

There is a significant impact on ocean productivity that arises from the change in aeolian Fe supply, with a reduction of up to 8% in the rate of uptake of anthropogenic CO₂ from the atmosphere (Figure 8c). This perturbation of the global carbon cycle exhibits a considerable persistence; the cumulative loss in the ocean carbon sink continues to increase after dust supply is stabilized in 2050 (Figure 8d). The deficit reaches $20-50\times10^9$ tons of carbon (or 20-50 PgC) by 2250, and perhaps doubling by the end of the millennium (year 3000). To put this into perspective, the potential sequestration benefit of widespread alteration of agricultural management practices and forestation is perhaps in the region of 23-110 PgC [*Royal Society*, 2001]. Clearly, suppression of the ocean sink has the potential to substantially offset the benefit to the atmosphere of sequestration on land.

Is a scenario for a global 15-30% reduction in future dust supply at all plausible? Results of early dust models suggested that a substantial (30-50%) component to present-day global dust supply originated in disturbed soils [*Tegen and Fung*, 1995]. Ameliorating the effect of past human-driven changes in land use could then potentially give rise to a 30% dust reduction. Recent satellite-based analysis now suggests a much smaller anthropogenic component, making a global change of this magnitude unlikely (although a substantial impact on dust mobilization is still discernable) [*Prospero et al.*, in press]. The precise effect on the global carbon cycle of the 'land use / ocean productivity' mechanism outlined here will be critically dependent upon the details of any sequestration activities and the locations in which they take place. For instance, the major dust sources are located in arid regions with annual rainfall less than 200-250 mm [*Prospero et al.*, in press] – areas of little agricultural activity. As such, one would not expect

these important sources to be directly influenced by future land use change. However, the Chinese government currently has firm plans for a massive reforestation program to combat soil erosion and associated dust storms in the loess region – this clearly has important implications for aeolian iron supply to the iron-sensitive equatorial and North Pacific. Thus, future changes in dust supply will probably occur on a regional scale rather than globally. Since socio-economic and political factors are likely to ultimately dictate such changes, future dust supply cannot be predicted with any certainty.

Although necessarily simplistic, these model results do serve to give an indication of the limit of maximum possible effect. The true value of this analysis, though, lies in the recognition of a hitherto overlooked causal link within the Earth system. At a minimum, changes in dust supply may need to be taken into account when evaluating the economics of carbon sequestration via certain LULUCF activities. However, it is within the range of uncertainty that the eventual benefit (in terms of reduced atmospheric xCO_2) obtained through implementation of LULUCF mitigation measures could be largely negated by an antagonistic response induced in the ocean. By not adopting an Earth system approach, but instead taking a rather narrow and restricted (land-atmosphere) view of the system, the validity of the carbon budgeting framework outlined in the Kyoto Protocol must be called into question.

5. The demise of the last ice age: a role for dust?

The Earth has experienced a series of intense ice ages over the course of the last million years or so. Each ice age has ended suddenly, with a rapid warming transition ('termination') from cold glacial conditions into a (relatively brief) mild interglacial period (Figure 9a). Many different theories have been advanced for how these cycles might be driven. These have typically focused on the physical climate system, particularly interactions between ice sheets and underlying bedrock (and forced by orbitally-modulated variations in the seasonal intensity of sunlight received at the Earth's surface). However, such explanations have not been able to account for

the magnitude and timing of the observed cyclicity in global ice volume, suggesting that some critical climatic factor outside of the physical system has been omitted [*Ridgwell et al.*, 1999].

Records of past atmospheric composition, in the form of microscopic bubbles of ancient air trapped within the crystalline structure of ice, has sparked a revolution in thinking regarding how these ice age cycles might be driven. Ice cores recovered from Greenland and Antarctica during the early 1980s and analyzed for air bubble gas composition revealed that atmospheric xCO_2 during the height of the last glacial was only about 190 ppm, compared with about 280 ppm before the start of the Industrial Revolution [*Houghton et al.*, 2001]. A core of over 3 km in length recovered from Vostok (in Antarctica) has been found to record over 420 thousand years (ka) of Earth history [*Petit et al.*, 1999]. Recent analysis of the CO₂ content of air bubbles contained in this core reveals a similar pattern to that of temperature, with relatively high atmospheric xCO_2 (~280 ppm) during interglacials, and low atmospheric xCO_2 (~190 ppm) during the most intense glacial periods (Figure 9b). This correlation with global climate has important implications for our understanding not only of how the global carbon cycle and climate system have interacted in the past, but also how the Earth system might respond over the next few centuries to the continued emission of CO₂ to the atmosphere [*Houghton et al.*, 2001].

What causes the observed variability in CO₂? A clue to the glacial-interglacial control of atmospheric xCO₂ comes from the observed changes in dust deposition, also recorded in the Vostok core (Figure 9c). The concentration of dust contained within the ice exhibits a series of rather striking peaks against a background of relatively low values; a much greater dynamic range than can be accounted for by dilution effects arising from changes in snow accumulation rate alone. The occurrence of these peaks correlates with periods of particularly low atmospheric xCO₂ values. Noting this correlation, and already suspecting that biological productivity in the Southern Ocean was Fe limited, John Martin hypothesized that enhanced dust supply to this region during the last glacial could have driven a more vigorous biological pump [*Martin*, 1990]. In this way, the low atmospheric xCO₂ values observed during glacial times might be explained.

Numerical models of the global carbon cycle that were subsequently developed have demonstrated that realistic increases in the strength of the biological pump in the Southern Ocean are unable to explain glacial atmospheric CO_2 mixing ratios as low as ~190 ppm (Figure 9b). However, of the total ~90 ppm deglacial rise in atmospheric xCO_2 , the initial 40-50 ppm occurs extremely rapidly (within just ~3 ka) and up to 10 ka before the collapse of the Northern Hemisphere ice sheets. Recent results from a carbon cycle model that explicitly accounts for the biogeochemical cycling of Fe in the ocean suggests that changes in the aeolian supply of Fe to the Southern Ocean (as indicated by the Vostok record) may be partly responsible for these particular features of the CO_2 record [*Watson et al.*, 2000]. Initial Fe-driven changes might be amplified by local feedbacks, perhaps through decreased sea ice extent or increased ocean surface temperatures to produce the entire 40-50 ppm of rapid rise. Obviously, to account for the remainder of the 90 ppm deglacial increase further mechanisms must be invoked once the ice sheets start to retreat [*Ridgwell*, 2001].

If dust is responsible for at least some of the observed glacial-interglacial variability in atmospheric *x*CO₂, what then drives the changes in dust flux? High glacial dust fluxes are not restricted to the Vostok site. There exist prominent common features in ice, marine, and terrestrial records of aeolian depositional from around the world. A colder, drier glacial climate, with a less vigorous hydrological cycle would result in decreased precipitation scavenging, more efficient transport of dust, and thus higher deposition rates. This is supported by models of dust generation, transport, and deposition run under the climatic conditions characteristic of glacial times [*Mahowald et al.*, 1999]. However, the predicted increases fall far short of observations. Although the confidence that can be placed in modelled dust deposition is much less than other environmental properties predicted by atmospheric general circulation models (such as surface temperature), deficiencies in model representation of transport and depositional processes cannot explain the magnitude of the model-data mismatch. Greater source strength of dust must therefore be invoked. For instance, with much of the continental shelves exposed during glacial periods as a result of lower sea levels, the land area available for dust production would be larger.

Glacial climates also favour the expansion of arid areas – preferential environments for the production and entrainment of dust [*Prospero et al.*, in press]. Greater source strength in conjunction with more efficient atmospheric transport can give rise to a substantial increase in dust deposition as compared to the present-day (Figure 10), and much closer to ice core data [*Mahowald et al.*, 1999].

6. Looking forward

We are just beginning to take a radical new 'defocused' view of how the Earth system functions on a range of timescales [*Schellnhuber*, 1999], one that is already bearing fruit. By considering the potential interaction of different system components, the intricate role that dust plays in biogeochemically linking land, air, and sea is starting to become apparent.

The transport of dust through the atmosphere provides a teleconnection between widely separated landmasses. If dust originating in North Africa exerts an important control upon biological productivity in Amazonia when deposited, an anti-phased relationship between the two regions is clearly possible. The aridity of North Africa has fluctuated widely in the past, with much greater vegetation cover being present about six thousand years ago. Could a more productive 'green' Sahel, by limiting dust supply, result in decreased productivity far away in the Amazon rainforest? Retrieval and analysis of marine pollen records recording vegetation changes in these two locations might provide a clue. As terrestrial vegetation and soil models are further developed to include multiple nutrient cycles and dust production processes, coupled soil-vegetation-climate models will also be useful in exploring such hypotheses.

With greater understanding of the causes and consequences of changes in dust supply, we may find that the role of dust is much more integral to the operation of the climate system than simply as a passive 'communicator' of events between components of the Earth system [*Ridgwell and Watson*, in press]. For instance, if changes in dust flux affect atmospheric xCO_2 (and thus climate), and dust fluxes are in turn responsive to global climate (such as through changes in sea

level and the strength of the hydrological cycle), this raises the possibility of feedback loops, as outlined in Figure 11. Any intensification in glacial state will tend to produce an increase in dust availability and transport efficiency. This could, in turn, produce a decrease in atmospheric xCO_2 (through Southern Ocean iron fertilization), causing a further intensification in glacial state and thus enhanced dust supply. Operation of this feedback loop would come to an end once the global carbon cycle has reached a second state, one in which biological productivity becomes insensitive to further increases in aeolian Fe supply, perhaps through the onset of limitation by NO₃⁻ [*Ridgwell and Watson*, in press; *Watson et al.*, 2000]. If aeolian Fe supply were then to decrease sufficiently to start limiting biological productivity again, the feedback loop operating in the opposite direction would act so as to reverse the original climatic change. That the Earth system might exhibit two distinct states, one of 'high- xCO_2 low-dust', and the other 'low- xCO_2 high-dust', is consistent with developing views of the climate system as being characterized by the presence of different quasi steady states with abrupt transitions between them.

To fully evaluate the role of feedbacks and linkages in the operation of the climate system, (such as are given rise to by dust), fresh investigative tools are required – numerical models of the Earth system. By coupling together representations of ocean and atmospheric circulation, cryosphere, and descriptions of the primary biogeochemical cycles that permeate the land, atmosphere, ocean, and sediments, the operation of the 'natural' climate system on a range of time scales can be explored. If these models are further extended by integration with socio-economic models, the interaction between the 'natural' system and anthropogenic activities can be addressed. Climate-socio-economic 'Integrated Assessment Models' are currently being actively developed by institutions such as the UK Tyndall Centre, and will greatly aid us in deciding how we might mitigate and adapt to future climate change.

The next few years are likely to see further dramatic discoveries regarding how the different components of the Earth system interact to govern the behaviour of the whole, and in doing so, determine the conditions for life on our planet.

AJR acknowledges support for this work from the IRONAGES project of the EU and from the Tyndall Centre for Climate Change Research.

References

- Boyd, P. W., *et al.* 2000 A mesoscale phytoplankton bloom in the polar Southern Ocean stimulated by iron fertilization. *Nature* **407**, 695-702.
- Conkright, M. E., Levitus, S. & Boyer, T. P. 1994 World Ocean Atlas 1994 Volume 1: Nutrients. NOAA Atlas NESDIS 1, U.S. Department of Commerce, Washington, D.C. 150 pp.
- Ginoux, P., Chin, M., Tegen, I., Prospero, J., Holben, B., Dubovik, O. & Lin S.-J. 2001 Global simulation of dust in the troposphere: Model description and assessment. *J. Geophys. Res.* 106, 20,255-20,273.
- Harrison, S. P., Kohfeld, K. E. Roeland, C. & Claquin, T. 2001 The role of dust in climate today, at the last glacial maximum and in the future. *Earth-Science Reviews* **54**, 43-80.
- Houghton, J. T., et al. 2001 Eds., Climate Change 2001: Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change, CUP, Cambridge, UK and New York, USA.
- Kennedy, M. J., *et al.* 1998 Changing sources of base cations during ecosystem development, Hawaiian Islands. *Geology* 26, 1015-1018.
- Mahowald, N., Kohfeld, K. E., Hansson, M., Balkanski, Y., Harrison, S. P., Prentice, I. C., Schulz, M. & Rodhe, H. 1999 Dust sources and deposition during the Last Glacial Maximum and current climate: A comparison of model results with paleodata from ice cores and marine sediments. *Journal of Geophysical Research*, **104**, 15895-15916.

- Martin, J. H. 1990 Glacial-interglacial CO₂ change: The iron hypothesis. *Paleoceanography* **5**, 1-13.
- Miller, R. L. & Tegen, I. 1998 Climate response to soil dust aerosols. *Journal of Climate* 11, 3247-3267.
- Petit, J. R., *et al.* 1999 Climate and atmospheric history of the past 420000 years from the Vostok ice core, Antarctica. *Nature* **399**, 429-436.
- Piketh, S. J., Tyson, P. D. & Steffen, S. 2000 Aeolian transport from southern Africa and iron fertilization of marine biota in the South Indian Ocean. *South African Journal of Science* 96, 244-246.
- Prospero, J. M., Ginoux, P., Torres, O., Nicholson S. & Gill, T. in press Environmental Characterization of Global Sources of Atmospheric Soil Dust Identified with the NIMBUS-7 TOMS Absorbing Aerosol Product. *Reviews of Geophysics*.
- Ridgwell, A. J. 2000 Climatic effect of Southern Ocean Fe fertilization: Is the jury still out? Geochem. Geophys. Geosys. 1. (http://g-cubed.org/)
- Ridgwell, A. J. 2001 Glacial-interglacial perturbations in the global carbon cycle, PhD thesis, Univ. of East Anglia at Norwich, UK. (http://tracer.env.uea.ac.uk/e114/ridgwell_2000.pdf)
- Ridgwell, A. J., Maslin, M. A. & Watson, A. J. 2002 Reduced effectiveness of terrestrial carbon sequestration due to an antagonistic response of ocean productivity. *Geophys. Res. Lett.* 29, 10.1029/2001GL014304.
- Ridgwell, A. J. & Watson, A. J. in press Glacial coupling between climate and CO₂: The Patagonian connection. *Paleoceanography*.

- Ridgwell, A. J., Watson, A. J. & Raymo, M. E. 1999 Is the spectral signature of the 100 kyr glacial cycle consistent with a Milankovitch origin? *Paleoceanography* **14**, 437-440.
- Royal Society 2001 "The role of land carbon sinks in mitigating global climate change". *Royal Society Document* **10/01**. (http://www.royalsoc.ac.uk/files/statfiles/document-150.pdf)
- Schellnhuber, H. J. 1999 'Earth system' analysis and the second Copernican revolution. *Nature* 402, C19-C23.
- Shinn, E. A., et al. 2000 African dust and the demise of Caribbean coral reefs. Geophysical Research Letters 27, 3029-3032.
- Swap, R., Garstang, M. Greco, S. Talbot, R. & Kallberg, P. 1992 Saharan dust in the Amazon Basin. *Tellus* 44, 133-149.
- Takahashi, T., Feely, R. A., Weiss, R. F., Wanninkhof, R. H., Chipman, D. W., Sutherland, S. C. & Takahashi, T. T. 1997 Global air-sea flux of CO₂: An estimate based on measurements of sea-air *p*CO₂ difference. *Proc. Nat. Acad. Sci. U.S.A.* 94, 8292-8299.
- Taylor, S. R., & McLennan, S. M. 1985 The continental crust, its composition and evolution: An examination of the geochemical record preserved in sedimentary rocks, Blackwell, Oxford.
- Tegen, I. & Fung, I. 1995 Contribution to the atmospheric mineral aerosol load from land-surface modification. *Journal of Geophysical Research* **100**, 18707-18726.
- Watson, A. J. 1997 Volcanic iron, CO₂, ocean productivity and climate. *Nature* 385, 587-588.
- Watson, A. J., Bakker, D. C. E., Ridgwell, A. J., Boyd, P. W. & Law, C. S. 2000 Effect of iron supply on Southern Ocean CO₂ uptake and implications for glacial atmospheric CO₂. *Nature* 407, 730-733.

Watson, A., Liss, P. & Duce, R. 1991 Design of a small-scale in situ iron fertilization experiment. *Limnol. Oceanogr.* 36, 1960-1965. Figure 1. True color satellite (SeaWiFS) image taken on February 26th 2000 of a massive sandstorm blowing off northwest Africa and reaching over 1000 miles into the Atlantic. (The SeaWiFS image was provided by NASA DAAC/GSFC and is copyright of Orbital Imaging Corps and the NASA SeaWiFS project.)

Figure 2. Model simulated distribution of the annual mean (1981-1997) rate of dust deposition to the Earth's surface [*Ginoux et al.*, 2001].

Figure 3. Schematic diagram of the operation of the 'biological pump' in the ocean. 'DIC' = (total) dissolved inorganic carbon $(CO_{2(aq)} + H_2CO_3 + HCO_3^- + CO_3^{2-})$. 'POM' = particulate organic matter (primarily living and dead phytoplantkon cells and zooplankton fecal pellets).

Figure 4. Global distribution of near-surface (30 m depth) ocean nitrate (NO₃⁻) concentrations [*Conkright et al.*, 1994].

Figure 5. Time series results of the SOIREE Fe fertilization experiment [*Boyd et al.*, 2000]. Empty and filled symbols represent measurements taken within and outside of the fertilized patch, respectively. **a**, Dissolved Fe concentrations in surface waters (at \sim 3 m depth), with the times of the four separate Fe infusions made during the course of the experiment indicated by arrows. **b**, Chlorophyll *a* density, integrated over the depth of the surface mixed layer (65 m).

Figure 6. Ocean color satellite (SeaWiFS) image of surface ocean chlorophyll *a* concentrations (cloud cover indicated by black regions), taken some 6 weeks after the deliberate release of iron in the Southern Ocean. By this time the fertilized ocean patch, centered on 61°S 141°E, was in the form of a ribbon ~100 km across. (SeaWiFS data provided by the NASA DAAC/GSFC and copyright of Orbital Imaging Corps and the NASA SeaWiFS project, and processed at CCMS-PML.)

Figure 7. Mean annual ocean-atmosphere CO_2 exchange in the South Indian Ocean [*Takahashi et al.*, 1997]. The plume trajectory taken by Fe-laden aerosols originating in southern Africa [*Piketh et al.*, 2000] is shown superimposed as a hatched region. Two prominent 'hot spots' of enhanced CO_2 uptake by the ocean can be seen to lie directly within the plume trajectory.

Figure 8. Model results of the effect of reduced aeolian iron supply on ocean carbon uptake [*Ridgwell et al.*, 2002]. **a**, Prescribed atmospheric xCO_2 history. **b**, Net annual (anthropogenic) carbon uptake, under 0% (solid line), 15% (dotted), and 30% (dashed) dust reduction scenarios. **c**, Suppression of anthropogenic carbon uptake compared to the baseline (i.e., 0% reduction) scenario. **d**, Cumulative suppression of the oceanic carbon sink.

Figure 9. Key indicators of climatic state contained within the Vostok ice core [*Petit et al.*, 1999]. **a**, Isotopically-derived temperature change (relative to the present) at the surface. Cold glacial and warmer interglacial ('IG') intervals are indicated. **b**, CO₂ concentration in air bubbles contained within the ice. **c**, Dust concentration in the ice. The correspondence between the occurrence of CO₂ minima and prominent dust peaks are highlighted.

Figure 10. Deposition predicted by a dust production, transport, and deposition model [*Mahowald et al.*, 1999]. **a**, Simulation assuming present-day climate and dust sources. **b**, Simulation assuming last glacial maximum climate and dust sources.

Figure 11. Schematic diagram of the hypothetical glacial dust-CO₂-climate feedback system [*Ridgwell and Watson*, in press]. Different components of the Earth system can directly interact in three possible ways; a positive influence (whereby an *increase* in one component directly results in an *increase* in a second – indicated by red arrows in the diagram), a negative influence (an *increase* in one component directly results in a *decrease* in a second – black arrows), or no influence at all. An even number (including zero) of negative influences occurring within any given closed loop gives rise to a positive feedback, the operation of which will act to amplify an initial perturbation. For instance, the 2-way interaction apparent between temperature and ice

23

volume is the 'ice-albedo' feedback. Conversely, an odd number of negative influences gives rise to a negative feedback, which will tend to dampen any perturbation. Primary interactions in the dust-CO₂-climate subcycle indicated by thick solid lines, while additional interactions (peripheral to the discussion here) are shown dotted for clarity. Four main (positive) dust-CO₂-climate feedback loops exist in this system; (1) dust supply—productivity—xCO₂—temperature—ice volume—sealevel—dust supply (4 negative interactions), (2) dust supply—productivity—xCO₂—temperature—hydrological cycle—vegetation—dust supply (2 negative interactions), (3) dust supply—productivity—xCO₂—temperature—hydrological cycle—dust supply (2 negative interactions), and (4) dust supply—productivity—xCO₂—temperature—hice volume—dust supply (2 negative interactions). Dust in the Earth system Dust in the Earth system

element	abundance (by mass)
0	44%
Si	31%
Al	8%
Fe	4%
Ca	3%
Κ	3%
Na	3%
Mg	1%
Р	1%

Table 1. Approximate abundance of the common elemental constituents of mineral dust (fromTaylor and McLennan [1985])











Figure 5







Figure 8



Figure 9





