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Feedback between aeolian dust, climate, and atmospheric CO_2 in glacial time

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[1] Enhanced aeolian supply of iron to the biota of the Southern Ocean during glacial periods is suspected to be an important contributory mechanism to the concurrently low observed mixing ratios of atmospheric CO_2. Declining rates of dust deposition prior to the glacial terminations may be critical in driving the initial deglacial rise in CO_2, but the reasons behind the dust decline itself are as yet unknown. Here we show that the dust record from the Vostok ice core can be qualitatively derived from a few general assumptions regarding the formation and aging of Patagonian sources of aeolian material and the efficiency with which it is transported through the atmosphere. We suggest that during glacial periods this dust supply becomes particularly sensitive to changes in global climate and that, in turn, climate is responsive to the dust due to iron fertilization. This positive feedback may mean that during glacial periods the carbon cycle exhibits two quasi steady states, characterized by distinct CO_2 concentrations. Recognition of this "glacial subcycle" may help to account for the timing and sequence of events at the terminations. *INDEX TERMS:* 1620 Global Change: Climate dynamics (3309); 3344 Meteorology and Atmospheric Dynamics: Paleoclimatology; 4267 Oceanography: General: Paleoceanography; 4805 Oceanography: Biological and Chemical: Biogeochemical cycles (1615); *KEYWORDS:* dust, CO_2, glacial, climate, Vostok

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1. Constraints Upon Aeolian Iron Supply to the Southern Ocean

[2] The global carbon cycle is likely to play a key role in the glacial-interglacial cycles of the late Quaternary [*Ridgwell et al.*, 1999; *Shackleton*, 2000]. Despite the fact that a full understanding of the observed variability in the atmospheric mixing ratio of CO_2 ($x$$CO_2$) recorded in ice cores [*Petit et al.*, 1999] still eludes us [*Archer et al.*, 2000b; *Ridgwell*, 2001; *Sigman and Boyle*, 2000], there is good reason to suspect that the decline in atmospheric dust loading that apparently proceeds each glacial termination [*Petit et al.*, 1999] may be linked to the initial deglacial rise in atmospheric $x$$CO_2$. The mechanism for this is "iron fertilization" of the biota in the Southern Ocean [*Watson et al.*, 2000]: the "iron hypothesis" [*Martin*, 1990]. However, it is far from universally accepted that enhanced aeolian Fe supply could lower atmospheric $x$$CO_2$ to any significant degree. Because there may be far-reaching implications of the Vostok dust record should there exist such a role, we first address recent criticisms made of the "iron hypothesis."

[3] The existence of a direct causal link between the decline in dust and the rise in atmospheric $x$$CO_2$ associated with each deglacial transition has been strongly argued against on the basis of an apparent lag in response of up to ~5 ka [*Broecker and Henderson*, 1998]. However, that a system as complex as the global carbon cycle might not respond linearly in this respect should not be entirely unexpected. In previous modeling work [*Ridgwell*, 2001; *Watson et al.*, 2000] we have demonstrated that a highly nonlinear response of atmospheric $x$$CO_2$ to increasing aeolian iron supply is consistent with the Vostok data [*Petit et al.*, 1999], and could arise through a number of factors; rapid onset of secondary (H_4SiO_4) limitation, increasing insensitivity of growth rates (Michaelis Menten saturation kinetics) [*Aksnes and Egge*, 1991] and decreasing efficiency of Fe use by phytoplankton [*Sunda and Huntsman*, 1995] (both a function of Fe availability), and the effect of loading [*Spokes and Jickells*, 1996] and "self-scavenging" on the effective solubility of aeolian Fe [*Ridgwell*, 2001]. Models predict a degree of variability may still be present even during peak glacial times [*Ridgwell*, 2001; *Watson et al.*, 2000], but because the amplitude of this response is highly dampened, an apparent lag is produced between the start of dust decline and any significant CO_2 change (as observed).

[4] More recently, *Maher and Dennis* [2001] dismiss the "iron hypothesis" on the basis that dust supply to high

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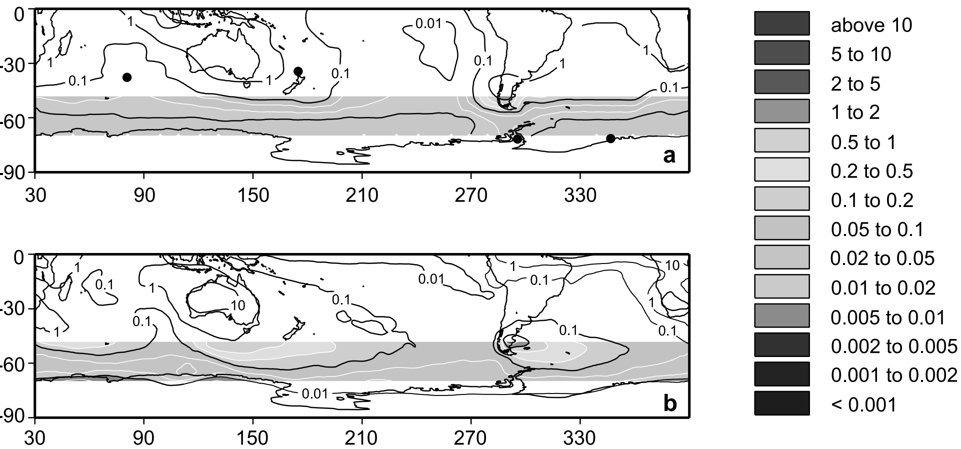

Figure 1. Observed [*Duce et al.*, 1991] (top) and model [*Mahowald et al.*, 1999] (bottom) estimated distribution of dust fluxes to the Southern Ocean (taken to be 47.5°S–70°S). Dust deposition is plotted as an annual mean (g m^{-2} a^{-1}). Also plotted on the Duce distribution are the locations (black dots) of the closest observational measurements made of atmospheric dust concentrations, on which the interpolated distribution is based [*Duce et al.*, 1991; *Jickells and Spokes*, 2001]. See color version of this figure at back of this issue.

Southern Hemisphere latitudes is always too low to significantly influence atmospheric xCO_2. In particular, they argue that the deposition field predicted by a dust availability-transport-deposition model [*Mahowald et al.*, 1999] and used to inform many current ocean carbon cycle models [e.g., *Archer and Johnson*, 2000; *Archer et al.*, 2000b; *Ridgwell*, 2001; *Ridgwell et al.*, 2002; *Watson et al.*, 2000] very substantially overestimates the present-day dust flux as compared with observational estimates [*Duce et al.*, 1991]. For instance, *Maher and Dennis* [2001] suggest that dust fluxes to the "Southern Ocean" in the carbon cycle model of *Watson et al.* [2000] (47.5°S–70°S) may be overestimated by 14–140 times, casting doubt upon the credibility of atmospheric xCO_2 predictions made by such models.

[5] The distributions of dust flux to the Southern Ocean estimated from data [*Duce et al.*, 1991] and dust availability-transport-deposition models [*Mahowald et al.*, 1999] in question are shown in Figures 1a and 1b , respectively. Although model-estimated isolines of dust flux appear to be shifted by up to 10–15° further to the south compared to the data, integrating the two dust maps over the region in question (47.5°S–70°S) we find a difference of a factor of about four. The much larger factor of 14–140 [*Maher and Dennis*, 2001] arises only when (arguably unrepresentative) point measurements are contrasted with an area-integrated mean. However, the paucity of the data used to delineate the interpolated distribution at these latitudes (Figure 1a) also limits the value of area-integrated comparison. Gridded estimates of the flux to the Southern Ocean used to inform ocean models, obtained from either data or by simulation, must therefore be treated with caution.

[6] In terms of understanding the operation of the carbon cycle in this climatically pivotal region, it is not absolute dust fluxes that are important, per se, but the supply of Fe in a dissolved form readily available to the biota. The solubility of iron in dust is very poorly constrained [*Jickells and Spokes*, 2001], with the result that this factor must often be treated in model studies as a "tunable" parameter; its value being chosen to achieve reasonable present-day ocean nutrient distributions [*Archer and Johnson*, 2000; *Archer et al.*, 2000b; *Ridgwell*, 2001]. Taking the *Mahowald et al.* [1999] dust deposition field as a boundary condition, solubility values for aeolian Fe in the Southern Ocean of 2% [*Ridgwell*, 2001; *Watson et al.*, 2000] or 6% [*Archer and Johnson*, 2000; *Archer et al.*, 2000b] have then been chosen. In contrast, field evidence suggests iron solubility in remote ocean regions can be as high as 10–50% [*Zhuang et al.*, 1990]. This is supported by recent solubility estimates (~10–90%) made in snow samples collected from coastal regions and seasonal sea ice in the Antarctic [*Edwards and Sedwick*, 2001]. It is therefore entirely possible that the rather low aeolian Fe solubilities assumed in models implicitly compensate for an overestimate in dust flux; the net result being a reasonable value for the flux of dissolved iron to the biota. Though there remains substantial uncertainty, the best current estimates of dissolved Fe input from dust input to the Southern Ocean appear compatible with model assumptions.

[7] An associated argument made against the "iron hypothesis" concerns the negligible contribution that aeolian material often appears to make to deep-sea sediments in the Southern Ocean [*Maher and Dennis*, 2001]. However, there is no a priori reason to expect that a dust flux sufficient to play a significant role in the carbon cycle of this region should necessarily leave a substantive signal in the sediment. To place the sedimentary contribution of dust into perspective, the sedimentary accumulation rate of biogenic opal (g m^{-2} a^{-1}) in an iron-limited situation can be crudely estimated as follows:

$$F_{SiO2} = 60 \cdot 0.05 \cdot 0.5 \cdot 3 \times 10^5 \cdot \frac{1}{x} \cdot \frac{1}{56} \cdot 0.1 \cdot 0.035 \cdot F_{dust} \approx \frac{28}{x} \cdot F_{dust}, \tag{1}$$

where F_{dust} is the dust flux (g m^{-2} a^{-1}) at the ocean surface (assumed equal to the accumulation rate in the sediments), and x is the molar fraction of the total dissolved Fe supply to the biota (including dissolved supply by upwelling and seasonal overturning), which originates directly from dust. The various factors in the calculation are: atomic mass of Fe and molecular mass of SiO_2 are 56 and 60 g mol^{-1}, respectively, dust comprises 3.5 wt% Fe with a solubility of 10% [*Duce and Tindale*, 1991], diatoms dominate new production, with opal export estimated by taking a C:Fe ratio of 3×10^5 [*Sunda and Huntsman*, 1995] typical of relatively iron-limited conditions, and a Si:C ratio of 0.5 [*Dugdale and Wilkerson*, 1998]. The overall (i.e., combined water column and sedimentary) preservation efficiency in deep-sea sediments of opal exported from the surface ocean mixed layer is assumed to be 5%, consistent with estimates made at 50°S and 53°S in the Indian sector of the Southern Ocean [*Pondaven et al.*, 2000].

[8] Models of the ocean iron cycle [*Ridgwell*, 2001; *Ridgwell et al.*, 2002; *Watson et al.*, 2000] suggest that the value of x characterizing the Southern Ocean may be about 0.09 (i.e., 9% of the total dissolved Fe supply to the biota is derived directly from dust). According to equation 1, SiO_2 would then be expected to accumulate in the sediments at over 300 times the mass rate of aeolian material. In the absence of other diluting solid components (such as $CaCO_3$), this represents an abundance for dust of just ~0.3 wt%. However, bottom water transport of detritus is known to be very important in the Southern Ocean [*Bareille et al.*, 1994; *Diekmann et al.*, 2000], with typical sediments in this region often containing between 11 and 56 wt% (on a $CaCO_3$-free basis) of detrital material [*Van Cappellen and Qiu*, 1997]. This suggests that the aeolian component would be massively diluted by re-suspended material, explaining why it is sometimes undetectable [*Diekmann et al.*, 2000]. Even if markedly different assumptions are made regarding export production and opal preservation or the degree of local sediment focusing, dust is still only likely to make a minor contribution to the total detrital flux, as observed [*Bareille et al.*, 1994; *Diekmann et al.*, 2000].

[9] Assuming that at the present-day, dust directly contributes ~9% of the total dissolved Fe flux to the biota, a glacial enhancement of dust by a factor ×10 could potentially support a doubling of the organic carbon export flux (in the absence of secondary limiting factors such as additional nutrient limitations). Although globally, aeolian deposition during glacial times was perhaps only 2–3 times greater than at present [*Harrison et al.*, 2001], changes in the hydrological cycle act so as to amplify the relative glacial-interglacial contrast in deposition at higher latitudes [*Andersen and Ditlevsen*, 1998; *Yung et al.*, 1996], with Antarctic ice core evidence suggesting up to a 20-fold flux increase [*Mahowald et al.*, 1999]. An order of magnitude increase in aeolian iron deposition to the Southern Ocean (sufficient to have a significant impact on atmospheric xCO_2) is then consistent both with a much lower global mean change in dust flux and with the dominance of bottom water detritus transport over dust deposition in the Southern Ocean.

[10] Finally, changes in dust deposition have an indirect effect on the flux of dissolved Fe to the euphotic zone which may be important, and which has not been previously recognized. Increases in dust deposition to the ocean surface will not only affect the supply to the biota of dissolved Fe coming directly from dust, but will lead to a greater flux of Fe to the ocean interior associated with exported biogenic material (either incorporated into cellular constituents or scavenged on particle surfaces) [*Ridgwell*, 2001]. Remineralization of this enhanced particulate Fe flux is very likely to lead to greater dissolved iron concentrations at depth. Subsequent transport of this water to the surface by upwelling/mixing will thus further enhance the supply of dissolved Fe to the biota, this time as an indirect effect of increased dust flux. This indirect effect could potentially double the net (direct + indirect) effect of changes in dust deposition, halving the enhancement in glacial dust supply required to support a doubling of biological export (i.e., from factor ×10 to just ×5). Interrogation of a global carbon cycle model [*Ridgwell*, 2001] confirms the plausibility of this effect. The codependence of "iron from below" on "iron from above" may help bridge the apparent divide between the original "iron hypothesis" of *Martin* [1990] and newly hypothesized controls by upwelled Fe [*Latimer and Filippelli*, 2001].

2. Glacial-Interglacial Control of Aeolian Deposition at Vostok

[11] If declining dust delivery at high Southern Hemisphere latitudes drives at least part of the initial rise in atmospheric xCO_2 observed at each glacial termination [*Watson et al.*, 2000], one might look further back along the causal chain of events for the initiator of the glacial terminations. Specifically, we must ask; what causes the decline in dust concentrations recorded in the Vostok ice core [*Petit et al.*, 1999] between glacial and interglacial time? This is an intriguing question in itself, regardless of the actual role of aeolian Fe supply in the carbon cycle of the Southern Ocean. Indeed, at the glacial terminations the decline in dust flux appears to precede any significant change in other climate proxies, leading local Antarctic temperature by some ~5 ka and the collapse of the Northern Hemisphere ice sheets by up to ~10 ka [*Broecker and Henderson*, 1998].

[12] A possible clue to the control upon dust supply at Vostok comes from an apparent association between periods of rapidly increasing foraminiferal $\delta^{18}O$ values (as recorded in marine sediment cores) and prominent peaks in dust concentration (Figures 2a and 2b). If the benthic $\delta^{18}O$ record were interpreted simply as a proxy for global ice volume, the association highlighted in Figure 2 would indicate a correlation between enhanced dust deposition and increasing global ice volume. Geochemical analysis has identified Patagonia as the provenance of the dust deposited in East Antarctica [*Grousset et al.*, 1992], consistent with transportation across the Southern Ocean by the prevailing winds [*Iriondo*, 2000]. Since most of the increase in land-based ice volume occurs in the Northern Hemisphere, how could such a $\delta^{18}O$-Patagonian dust link arise? Climate across much of the two hemispheres often appears to change in synchrony, with Northern Hemisphere cooling (driven by the relatively high albedo of ice cover) [*Broccoli and Manabe*, 1987] propagated globally, perhaps through changes in the CO_2 and water vapor content of the atmos-

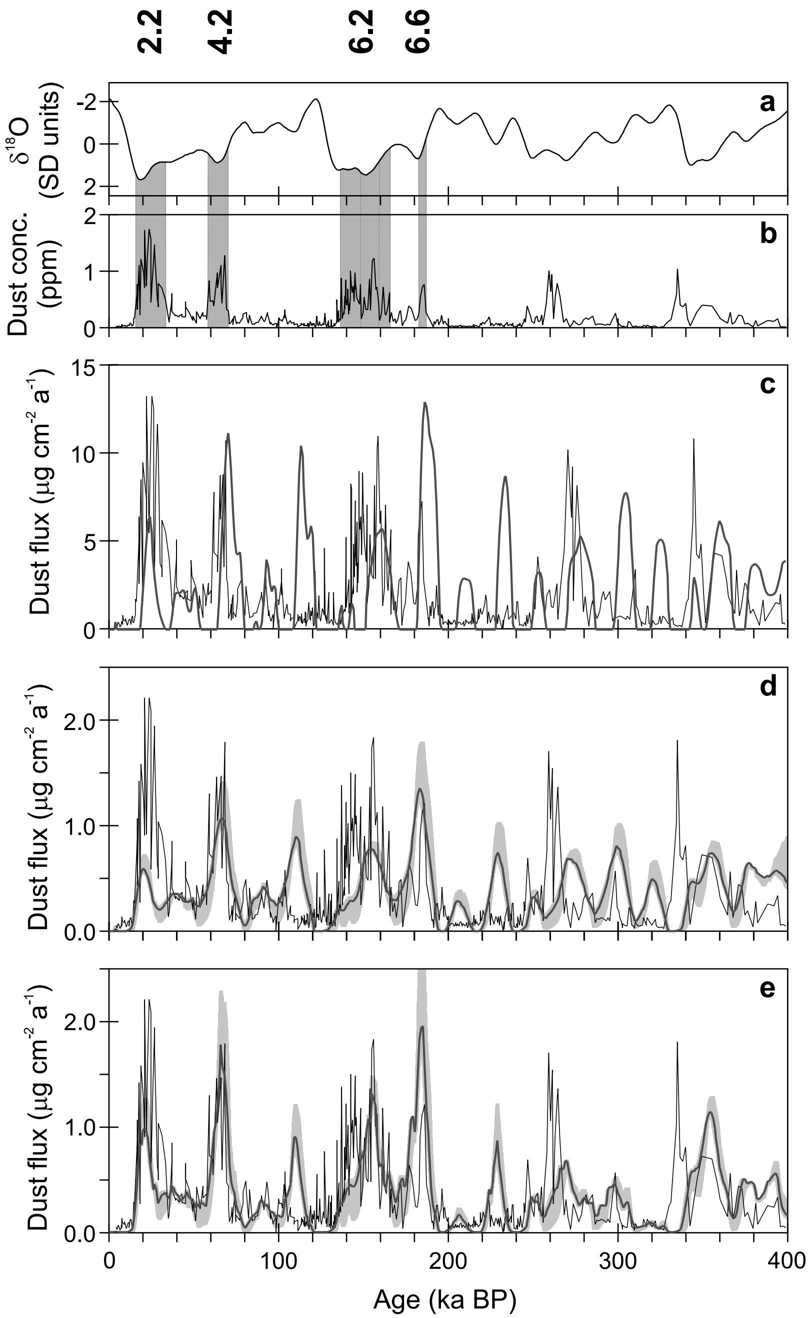

Figure 2. Observed and modeled relationship between dust and climate. (a) The SPECMAP $\delta^{18}O$ stack [*Imbrie et al.*, 1984] (which by convention is shown with the $\delta^{18}O$ scale reversed) with Marine Isotope Stages [*Bassinot et al.*, 1994] marked. (b) The Vostok dust concentration record [*Petit et al.*, 1999]. The apparent correspondence between prominent maxima in dust concentrations (Figure 2b) and periods of rapid increase in $\delta^{18}O$ (Figure 2a) during glacial times (loosely delineated by $\delta^{18}O$ greater than about 0.0 SD units) is shown highlighted (gray shading). In contrast to the rather close correlation over the earlier half of the record (0 to ~200 ka BP), there is no obvious analogous relationship between the two records over the older portion (~200 to 400 ka BP). (c) The Vostok dust depositional record (black line) reconstructed from dust concentrations [*Petit et al.*, 1999] and estimated snow accumulation history (J. B. Petit, personal communication, 2001) alongside the results of a simple (positive truncation) first-derivative model (red line). (d) Observations (black line) contrasted with the results of the initial source-only model (equation 3) (red line) with τ = 12.5 ka. Results from a sensitivity analysis with τ in the range 5–15 ka are marked in orange. (e) Observations (black line) contrasted with the results of the complete source + transport model (equation 4) (red line). Results from a sensitivity analysis with τ in the range 5–15 ka are marked in orange. See color version of this figure at back of this issue.

phere [*Cuffey and Vimeux*, 2001; *Lowell et al.*, 1995]. For example, events surrounding the most recent deglacial transition in central and southern Chile are found to be in phase with events in the Northern Atlantic region [*Clapperton*, 1998; *Lowell et al.*, 1995; *Moreno et al.*, 2001]. It is therefore reasonable to expect that increases in global ice volume may be coincident with a cooling climate in Patagonia. The relation between the Vostok dust record and the oxygen isotopic signature of the ocean would then reflect this correlation.

[13] We suggest two main mechanisms whereby periods of climatic cooling might give rise to significantly enhanced availability of Patagonian loess and thus increased deposition rates at Vostok. The first envisages that changes in dust source strength may be related to the production of fines (such as rock flour) by glacial erosion, in conjunction with the supply of this material to areas from where deflation can take place. With a cooling climate there will be an advance of the Patagonian ice cap, an increase in total erosion rate and thus enhanced supply of flavioglacial outwash material. However, total erosion rates would be expected to decline once newly extended glaciers reach their maximum extent and begin to adopt an equilibrium (“U-shaped”) profile [*Harbor*, 1992]. Additionally, with each successive glacial advance, soil material formed under earlier periods of temperate climate [*Pye*, 1989] would be rapidly eroded and soon depleted, also giving rise to a relaxation of the initial supply increase. That a first-derivative type relationship should exist between periods of rapid climatic change and peak erosion rates finds support in recent analyses of global sediment supply over the last ~80 Ma, in which enhanced erosion appears to be more closely associated with periods of climatic transition than the absolute level of any particular steady state [*Peizhen et al.*, 2001]. The second, but certainly not mutually exclusive, possibility, is based on the reduction in the intensity of the hydrological cycle that would result from a cooling of global climate [*Harrison et al.*, 2001] (which follows from the Clausius-Clapeyron relationship) [*Rind*, 1986]. New sources of dust are then created by a progressive aridification of Patagonia [*Iriondo and Garcia*, 1993]. These sources may not last indefinitely, and might decline slowly due to the gradual depletion of fine silty fractions and the eventual armoring and stabilization of the soil surface [*Pye*, 1989; *Rea*, 1994], although the timescale on which this process might operate is not clear.

[14] There is also a third possibility: interpreting the benthic $\delta^{18}O$ record as a proxy for sea level [*Shackleton and Opdyke*, 1973]. In this case, the correlation observed in Figure 2 would suggest that falling sea level in glacial time is associated with high dust concentrations in Antarctic ice. Incremental falls in sea level will result in increased exposure of the extensive Patagonian continental shelf. Because of the arid conditions of this region during glacials [*Iriondo and Garcia*, 1993], we would not expect the newly exposed shelf to become extensively vegetated. There is then the potential for the relatively unconsolidated marine sediments to be rapidly deflated by wind action. If the material available for deflation tends to “blow out” after a time, this would explain why the decline in dust flux begins in advance of sea-level rise at deglaciation [*Broecker and Henderson*, 1998]. However, recent geochemical analysis points to Patagonian loess rather than the shelf sediments themselves as the primary dust source [*Basile et al.*, 1997]. This would suggest that only a limited role could have been played by this “sea-level” scenario.

[15] Many of these arguments also predict a global component to dust variability since they are applicable over much of the earth and not just to Patagonia. This is supported by the existence of prominent common features in ice, marine, and terrestrial aeolian depositional records from around the world [*Kohfeld and Harrison*, 2001]. However, we restrict discussion to the potential link between Patagonia and the Antarctic region.

[16] We now construct a mathematical formulation to explore the link between the Vostok dust flux record and the $\delta^{18}O$ curve. All the mechanisms for dust generation described above predict that sources are created during times of climate cooling (positive-going changes in $\delta^{18}O$) and destroyed when climate warms (negative-going changes in $\delta^{18}O$). Taking into account the apparent correlation highlighted in Figures 2a and 2b, a direct first-derivative relationship between $\delta^{18}O$ and dust is an obvious possibility. Figure 2c shows the first derivative (positive truncation) of $\delta^{18}O$ compared with observed dust deposition, and scaled to have the same mean flux over the past 400 ka as the observed record. It can be seen that although peaks are correctly predicted associated with Marine Isotope Stages (MIS) 2.2 and 4.2 (adopting the revised scheme of *Bassinot et al.* [1994]), dust decline occurs too early in each instance (among other obvious mismatches). However, all the possible causal mechanisms discussed above suggest a degree of persistence of newly created dust sources, but falling short of an indefinite lifetime. We therefore make the additional assumption that the strength of dust sources decays with time, even in the absence of a climatic reversal. Incorporating these general features into a suitable algorithm, the total deflation source strength (in arbitrary units) at time t is expressed

$$S_{(t)} = e^{\frac{-\Delta t}{\tau}} \cdot S_{(t-\Delta t)} + H\left(\delta^{18}O_{(t)} - \delta^{18}O_{(t-\Delta t)}\right) \cdot \frac{\delta^{18}O_{(t)} - \delta^{18}O_{(t-\Delta t)}}{\Delta t} - H\left(\delta^{18}O_{(t-\Delta t)} - \delta^{18}O_{(t)}\right) \cdot \Delta s_{(t)}, \quad (2)$$

where Δt is the time step, τ is the source lifetime, H is the heavyside function, and $\Delta S_{(t)}$ accounts for the loss of dust sources with any amelioration in global climate. Choosing an appropriate scaling constant a, the depositional flux, F, can be written

$$F_{(t)} = a \cdot S_{(t)}. \quad (3)$$

We constrain the value of a by the requirement that the mean dust flux over the last 400 ka should be equal to the observed value (~0.37 $\mu g\ cm^{-2}\ a^{-1}$). The results of this algorithm are shown in Figure 2d. Despite the simplicity of the scheme, some of the general features of the Vostok record are reproduced, such as the prominent peaks associated with MIS 2.2, 4.2, 6.2, and 6.6. Crucially, there is some agreement between model and observations with respect to the timing of the rising and falling edges of these peaks. We find a reasonable value for the decay constant τ to be around 12.5 ka, although there is scant quantitative evidence with which

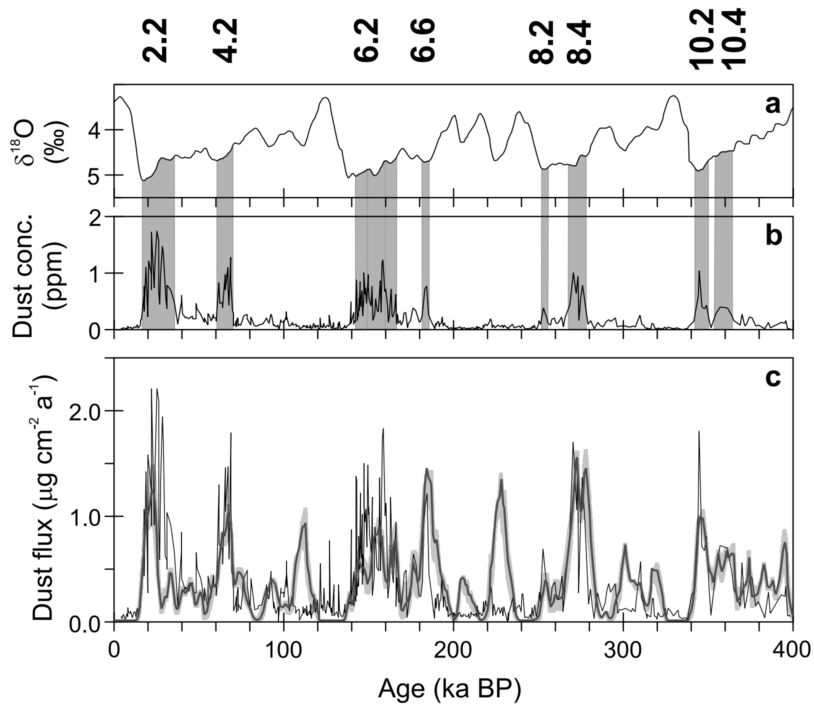

Figure 3. Observed and modeled relationship between dust and climate. (a) The benthic foraminiferal δ^{18}O record from V19-30 [*Shackleton*, 2000]. (b) The Vostok dust concentration record [*Petit et al.*, 1999], repotted on the same age scale as the marine δ^{18}O signal [*Shackleton*, 2000]. The apparent correspondence between prominent maxima in dust concentrations (Figure 3b) and periods of rapid increase in δ^{18}O (Figure 3a) is highlighted (gray shading). (c) Observations (black line) contrasted with dust fluxes (red line) predicted by the source + transport model (equation 4) with τ = 7.5 ka and taking the alternative δ^{18}O record as an input. Results from a sensitivity analysis with τ in the range 5–15 ka are marked in orange. See color version of this figure at back of this issue.

to guide our choice in this. However, we note that this value is comparable with the adjustment time ($\sim$5–19 ka) of the bedrock erosional profile (and thus of erosion rates) of a glacial system at steady state with respect to ice flow (i.e., one at its greatest extension, corresponding to a local maximum in δ^{18}O) [*Harbor*, 1992]. This is consistent with an important direct role for the Patagonian ice cap in the dust system of this region. What the inclusion of τ in the algorithm (2) achieves is to create an apparent lag between predicted dust decline and the first derivative of δ^{18}O sufficient to match the observed decline. It should be noted that τ is distinct from the response time of either ice sheets (which drive much of the δ^{18}O variability) or Andean glaciers (which will tend to follow climatic changes with a much shorter adjustment time of 10s to 100s of years). Despite the poorly constrained nature of τ, the sensitivity analyses conducted with τ in the range 5–15 ka (see Figure 2) suggest that the phase and amplitude of the model response is not particularly sensitive to this variable.

[17] The availability of material suitable for deflation is, of course, just the first necessary step toward its eventual deposition. This material must be entrained by winds of sufficient intensity and be transported considerable distances across the open ocean without first being scavenged by precipitation. As a result, the supply of dust to high latitudes will tend to be relatively inefficient in a warm, wet interglacial-type climate, but become progressively more favorable as the hydrological cycle weakens [*Andersen and Ditlevsen*, 1998; *Yung et al.*, 1996]. This is suggested visually by the lack of prominent dust maxima when δ^{18}O is low (Figure 2). That rapid increases in δ^{18}O associated with glacial inception (e.g., around 115 ka BP) are not correlated with a significant dust peak may then be because the hydrological cycle is too vigorous for efficient long-range dust transport at these times. A scalar modification of dust availability, υ, is now introduced into the algorithm, in an attempt to account for variability in the efficiency of atmospheric transport to Vostok. Assuming a key role for the hydrological cycle, a record of past changes in precipitation at Vostok is used as a basis for this function (formed by taking the reciprocal of the precipitation rate). We use the reconstructed snow accumulation rate of the Vostok core (J. P. Petit, personal communication, 2001), although a simple exponential relationship of past variation in precipitation with local temperature change [*Ritz et al.*, 2001] gives essentially the same function. A power, β, is applied to represent nonlinear scaling of transport efficiency with the intensity of the hydrological cycle [*Andersen and Ditlevsen*, 1998; *Yung et al.*, 1996]. The depositional flux is now

$$F_{(t)} = \upsilon_{(t)}^{\beta} \cdot a \cdot S_{(t)}, \tag{4}$$

the results of which are shown in Figure 2e. It can be seen that at least for the first $\sim$200 ka, the algorithm now produces a fair simile of the Vostok flux record. The exact form of the function υ is relatively unimportant, and the main results of this exercise are little changed should an artificial sawtooth with a mean period of 100 ka [*Ridgwell et al.*, 1999] be employed in its place. The main purpose it serves is to modify relative peak heights (e.g., enhancing the MIS 2.2 peak relative to that at MIS 4.2) and the form of the underlying background signal. A value $\beta = 3$ is adopted on the basis of the observed relationship between the GRIP dust concentration and accumulation rate record [*Andersen and Ditlevsen*, 1998; *Steffensen*, 1997] and assuming that that changes in transport efficiency dominate the observed variability over the last glacial cycle [*Fuhrer et al.*, 1999; *Svensson et al.*, 2000]. The results are also little changed if a slightly stronger role for the hydrological cycle were to be assumed (for instance, taking $\beta = 4$). In contrast to the earlier part of the record, the temporal correlation between periods of glacial intensification and maximum dust flux over the interval ~200 to 400 ka BP is poor. The chronologies for the input and target signals, however, are derived by very different methodologies; the SPECMAP $\delta^{18}O$ stack being tuned to an orbital template [*Imbrie et al.*, 1984], while the GT4 timescale employed for the Vostok core is glaciologically derived [*Petit et al.*, 1999]. There is, therefore, the potential that a loss in coherency between the two timescales might arise, one in which the degree of offset is exacerbated with increasing age. The analysis is now repeated employing chronologies derived via a common methodology [*Shackleton*, 2000] and taking as input the benthic foraminiferal $\delta^{18}O$ record from the equatorial Pacific core V19-30 [*Shackleton*, 2000] in place of the SPECMAP stack. This results in substantially better model-data agreement throughout the older portion of the record (Figure 3), to the extent that the split dust peak associated with MIS 8.4 is resolved and much of the variability in the interval 340–400 ka BP well reproduced.

[18] One obvious caveat to our dust flux predictions concerns the choice of marine $\delta^{18}O$ record used to drive the conceptual model. It can be seen (Figures 2e and 3c) that the fine-scale detail of the predicted dust flux is sensitive to the choice of this record. However, since changes in ice volume produce a global $\delta^{18}O$ effect [*Shackleton*, 2000], certain common features can be expected across marine records; for instance, local $\delta^{18}O$ maxima at MIS 2.2 and 4.2 [*Imbrie et al.*, 1984; *Raymo*, 1997]. It is these prominent (global) $\delta^{18}O$ maxima that appear to be associated with the major dust peaks. The primary features of the synthetic dust record will therefore be relatively insensitive to the choice of isotope record, and can be considered to be a robust result of our model. Results of additional experiments (not shown) carried out using alternative $\delta^{18}O$ records (such as core MD900963) [*Bassinot et al.*, 1994] as input to the model confirm this.

[19] It is worth noting that none of the dust models, even when the relative inefficiency of atmospheric transport during warm climates is taken into account, can successfully reproduce dust fluxes associated with glacial inception. This is particularly apparent immediately following MIS 5.5 and 7.5; while the dust model predicts large dust peaks driven by the initial rapid $\delta^{18}O$ increase, dust deposition as recorded at Vostok remains low. We suspect that the climate system is far more nonlinear than is accounted for in the model, such that there is no noticeable response of dust to cooling until a sufficiently glacial state of the climate system has been achieved.

3. Implications for Glacial Climate Dynamics

[20] We have recently shown [*Watson et al.*, 2000] that, during glacial periods (including the onset of terminations, but specifically excluding interglacials) it is possible to explain the close relation between atmospheric xCO_2 concentrations and dust fluxes as an effect of Southern Ocean iron fertilization. Assuming that the Vostok dust record can be taken as a proxy at least for the timing of changes in dust supply to the Southern Ocean, the observations discussed above suggesting that dust fluxes are in turn responsive to global climate change raises the possibility of a positive feedback. This is a system in which a change in one component causes a chain of effects having the net effect of an amplification of the initial change. A well-established example of this in the Earth system is the "ice-albedo" feedback [*Budyko*, 1969; *Gildor and Tziperman*, 2000, 2001], whereby an increase in land or sea ice cover increases surface albedo, resulting in a reduction in absorbed solar energy and a surface cooling, thus driving a tendency for a further increase in ice cover.

[21] We hypothesize that once a sufficiently "glacial" state of the climate system has been achieved, perhaps representing a critical balance of Fe supply to the surface Southern Ocean between aeolian delivery and upwelling from the deep ocean, the system becomes highly susceptible to perturbation. Any further glacial intensification will tend to result in an increase in the strength of dust sources and transport efficiency through the atmosphere. This will, in turn, produce a drawdown in atmospheric xCO_2 through Southern Ocean iron fertilization, causing a further intensification in glacial state and thus enhanced dust supply. Operation of this positive feedback loop (shown schematically in Figure 4a) will 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 [*Ridgwell*, 2001; *Watson et al.*, 2000] (as discussed earlier). If aeolian Fe supply then decreases sufficiently to start limiting biological productivity, the positive feedback loop will tend to reverse the original climatic change. A reduction in the strength of the biological pump and rising atmospheric xCO_2 leads to a warming climate with weaker dust sources and less efficient atmospheric transport, further decreasing the dust supply. In this way, the system will return to the initial state, with biological productivity relatively insensitive to further decreases in aeolian Fe supply to the Southern Ocean. Because our model investigations suggest that the initial supply rate of material associated with newly created dust sources cannot be indefinitely sustained (indicated by the presence of the decay constant τ in equation 2), the lower climatic state is intrinsically unstable, and the return transition will eventually occur even in the absence of a second (external) perturbation. Thus a transient climatic state characterized by high dust fluxes and low atmospheric xCO_2

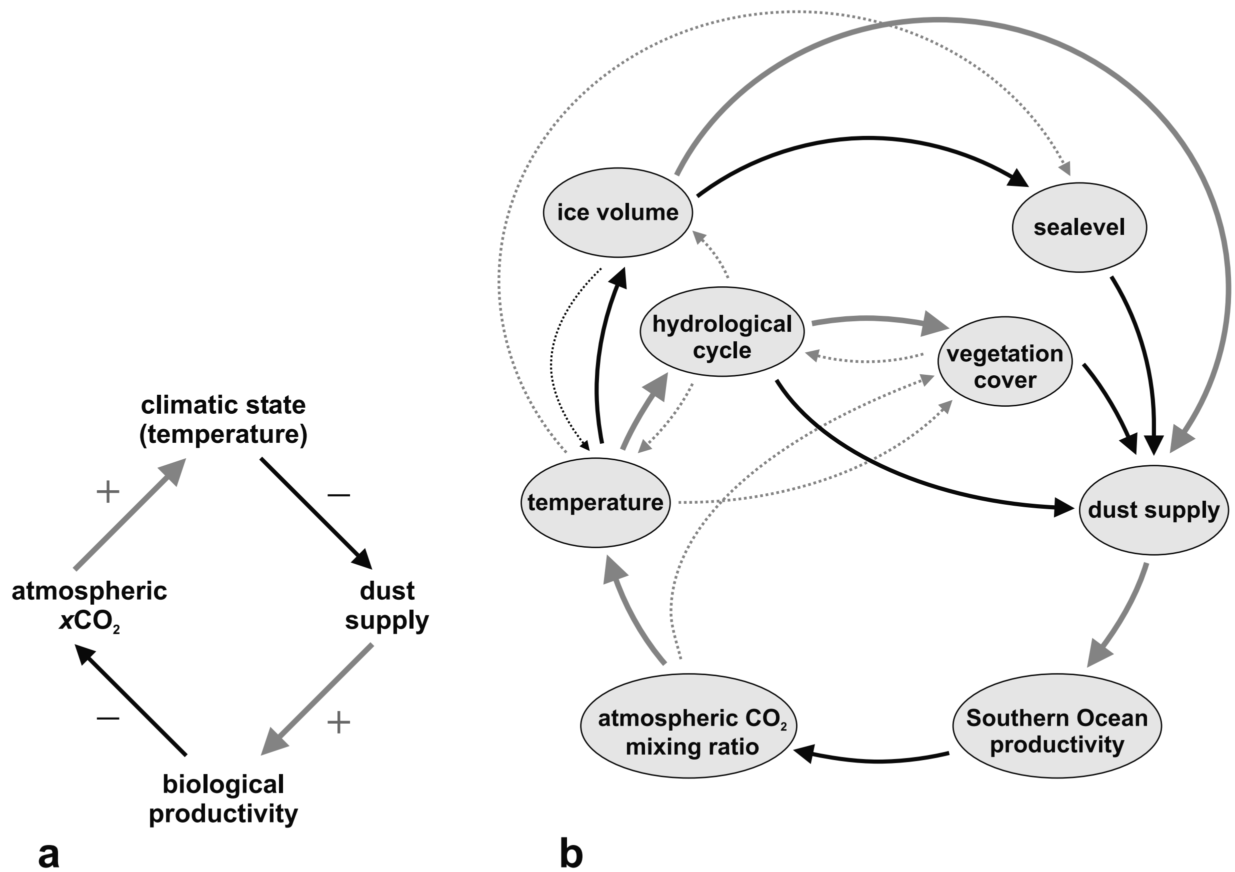

Figure 4. Feedbacks in the climate system. Different components of the Earth system can directly interact in three possible ways; a positive influence (i.e., an increase in one component directly results in an increase in a second), a negative influence (i.e., an increase in one component directly results in a decrease in a second), 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. Conversely, an odd number of negative influences gives rise to a negative feedback, which will tend to dampen any perturbation. In the two schematics, positive influences are shown in gray, and negative in black. (a) Schematic diagram of a simplified feedback system, involving dust, the strength of the biological pump, CO_2, and climatic state (represented by mean global surface temperature). Because there is an even number of negative influences (=2), this represents a positive feedback, with the potential to amplify an initial perturbation (in either direction). (b) Schematic diagram of our hypothetical glacial dust-CO_2-climate feedback system, with explicit representation of the various dust mechanisms that we have identified. Primary interactions in the dust-CO_2-climate subcycle are indicated by thick solid lines, while additional interactions (peripheral to our argument) are shown dotted for clarity. For instance, the 2-way interaction between temperature and ice volume is the "ice-albedo" feedback. Four main (positive) feedback loops exist in this system: (1) dust supply → productivity → xCO_2 → temperature → ice volume → sea level → dust supply (4 negative interactions), (2) dust supply → productivity → xCO_2 → temperature → hydrological cycle → vegetation → dust supply (2 negative interactions), (3) dust supply → productivity → xCO_2 → temperature → hydrological cycle → dust supply (2 negative interactions), and (4) dust supply → productivity → xCO_2 → temperature → ice volume → dust supply (two negative interactions).

might be generated from a single triggering event: the climatic equivalent of a monostable pulse generator in electronic engineering theory. Individual component interactions within the various possible permutations of our dust-CO_2-climate feedback system are shown schematically in Figure 4b.

[22] The existence of such a subsystem should not be unexpected. The emerging view of the climate system is characterized by the presence of different quasi steady states [*Gipp*, 2001; *Paillard*, 1998] together with abrupt transitions between them [*Paillard*, 2001], well-known (model) examples being the multiple states exhibited by the Atlantic overturning circulation [*Broecker et al.*, 1985; *Strommel*, 1961] and the Sahel atmosphere-vegetation system [*Claussen*, 1998; *Claussen et al.*, 1999]. The two distinct states in the glacial carbon cycle predicted by our dust-CO_2-climate

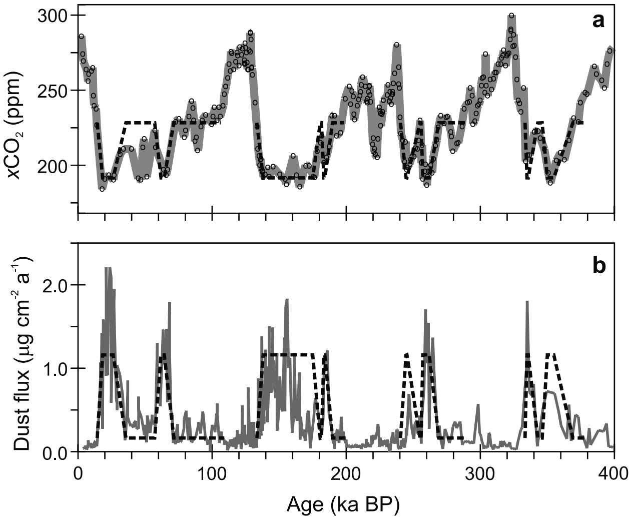

Figure 5. Conceptual operation of the global carbon cycle during glacial periods (i.e., specifically excluding interglacials). (a) The hypothetical history of transitions between system states suggested by the dust-CO_2-climate subcycle is indicated (black dashed line) compared to the Vostok CO_2 record [*Petit et al.*, 1999] (gray line with data points). Transitions from the lower to upper state appear to be generally faster than the reverse transition, consistent with the differential timescales characterizing the decay and growth of ice sheets [*Imbrie and Imbrie*, 1980]. (b) Associated hypothetical transitions in dust flux (black dashed line) compared to the observed record (gray line).

subcycle hypothesis are consistent with such a view, and are supported by apparent clustering at glacial CO_2 concentrations of ~190 and 225 ppm in $x$$CO_2$-$\delta^{18}$O space [*Saltzman and Verbitsky*, 1994]. To a first order, the intraglacial behavior of atmospheric $x$$CO_2$ recorded at Vostok [*Petit et al.*, 1999] as well as the general features of the dust record, is of residence in one of two distinct states, or in transition between them, as highlighted in Figure 5. For the two (out of the possible four) feedback loops we identify for which Northern Hemisphere ice volume is a key component, the transit times between states will be set by the differential timescales characterizing the decay and growth of ice sheets [*Imbrie and Imbrie*, 1980]. Transition times for alternative positive feedbacks involving the hydrological cycle could be much quicker, however (perhaps hundreds of years or less).

[23] Highly simplified "box" models of the ocean carbon cycle [e.g., *Broecker and Peng*, 1986; *Keir*, 1988] have recently been criticized on the grounds that the atmospheric $x$$CO_2$ sensitivity they exhibit in response to perturbation of the high latitude ocean surface (such as would be produced via the "iron hypothesis") might be overstated [*Archer et al.*, 2000a; *Broecker et al.*, 1999]. More complex models suggest that the direct effect (i.e., before the lagged response driven by $CaCO_3$-compensation) [*Broecker and Peng*, 1987] of glacial-interglacial changes in aeolian Fe supply might be around 22–34 ppm [*Archer et al.*, 2000b; *Kurz and Maier-Reimer*, 1993; *Ridgwell*, 2001]. Although this is slightly lower than the ~35 ppm estimated by *Watson et al.* [2000], the reasons for the intermodel differences in atmospheric $x$$CO_2$ sensitivity are not entirely clear [*Ridgwell*, 2001], and it is not yet possible to identify any particular model as having the "correct" response. However, initial Fe-driven changes in atmospheric $x$$CO_2$ are likely to be amplified by further local feedbacks, such as through changes in sea ice extent [*Stephens and Keeling*, 2000], ocean surface temperatures, and stratification [*Sigman and Boyle*, 2000]. Thus the operation of the dust-CO_2-climate subcycle could potentially account for the full ~35–40 ppm of intraglacial variability that is observed [*Petit et al.*, 1999], even if decreasing aeolian Fe supply directly drives less than 35 ppm of this. Obviously, further mechanisms must be invoked at the terminations to account for the remainder of the observed glacial-interglacial amplitude of atmospheric $x$$CO_2$ [*Archer et al.*, 2000b; *Ridgwell*, 2001]. Model studies suggest that once ice sheets are sufficiently old and massive, the long relaxation time of the continental bedrock makes them highly susceptible to melting perturbation and rapid deglaciations [*Oerlemans*, 1991]. Models also suggest that particularly low atmospheric $x$$CO_2$ (of order 210 ppm) is required for the growth of a sufficiently large ice sheet [*Berger et al.*, 1998]. By depressing $x$$CO_2$ by ~35 ppm, the dust-CO_2-climate subcycle we have described may then play a key role in bringing the ice sheet-bedrock system to a critical state [*Paillard*, 1998, 2001]. However, since the "low-$x$$CO_2$ high-dust" is inherently unstable, the system will tend to transit back to a "high-$x$$CO_2$ low-dust" state. The global climate warming caused by this rise in atmospheric $x$$CO_2$ may therefore help determine the timing of deglaciations, and this timing would not be tied precisely to any particular phase of the external, orbital forcing. A single triggering of the dust-CO_2-climate subcycle could, in theory, sequentially provide both the climatic conditions necessary for the growth of a massive ice sheet, and subsequently those necessary for its demise.

4. Conclusions

[24] By recognizing the potential link between the availability of dust in Patagonia and global climate change, we have been able to devise a simple theoretical framework that can account for the main features of the Vostok dust record [*Petit et al.*, 1999]. This hypothesis represents the first explanation that we are aware of for the timing of the dramatic decline in dust that immediately precedes each glacial termination. In seeking to elucidate linkages between the timing of events delineated within marine sediment cores and those delineated within ice cores, the value of a common chronology applicable to both mediums is apparent. In this regard, use of the elegant methodology for timescale generation devised by *Shackleton* [2000] clearly warrants more widespread consideration in future. Recent influential arguments have been made against the "iron hypothesis" [*Martin*, 1990], based upon the extremely low dust fluxes observed in the Antarctic, together with the negligible contribution that aeolian material appears to make to deep-sea sediments (both at present and during glacials) [*Maher and Dennis*, 2001]. However, we have shown that such arguments are overstated. The key consideration is not the absolute value of dust flux per se, but the

balance of dissolved Fe supply to the biota between aeolian deposition and upwelling. Without explicit reference to this balance, the simple observation that dust deposition rates to the Southern Ocean are among the lowest anywhere in the present ocean, by itself, can tell us nothing new. We show that a significant role for aeolian Fe in influencing atmospheric xCO_2 at the glacial terminations is not inconsistent with low dust deposition rates and the dominance of bottom water detrital transport over dust deposition in this region. However, there is clearly a pressing need to better quantify the operation of the oceanic iron cycle in climatically important regions such as the Southern Ocean (particularly in terms of such "basic" parameters as dust flux and iron solubility) as a prerequisite to furthering our understanding of the mechanisms behind observed paleoclimatic change. New studies carried out on snow samples taken from near-coastal Antarctic sites [*Edwards et al.*, 1998; *Edwards and Sedwick*, 2001] and from the sea-ice surface further to the north [*Edwards and Sedwick*, 2001] will be invaluable in this respect.

[25] If past changes in aeolian iron supply to the Southern Ocean have played some nontrivial role in determining the observed atmospheric xCO_2 record and thus affected climate, that changes in climate may, in turn, affect dust supply, results in a positive feedback loop. The prediction of two distinct states in the glacial carbon cycle; our "dust-CO_2-climate subcycle" that arises from this feedback, may prove to be invaluable in understanding the dynamics of the glacial climate system.

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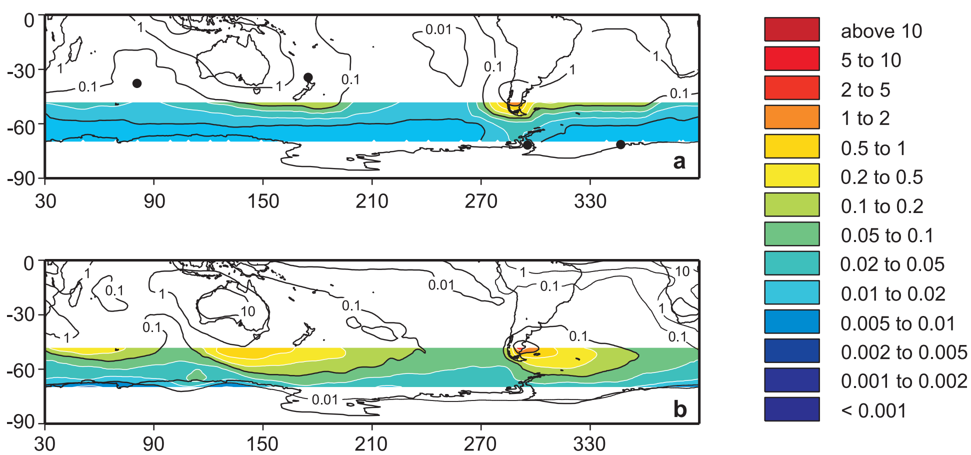

Figure 1. Observed [*Duce et al.*, 1991] (top) and model [*Mahowald et al.*, 1999] (bottom) estimated distribution of dust fluxes to the Southern Ocean (taken to be 47.5°S–70°S). Dust deposition is plotted as an annual mean (g m^{-2} a^{-1}). Also plotted on the Duce distribution are the locations (black dots) of the closest observational measurements made of atmospheric dust concentrations, on which the interpolated distribution is based [*Duce et al.*, 1991; *Jickells and Spokes*, 2001].

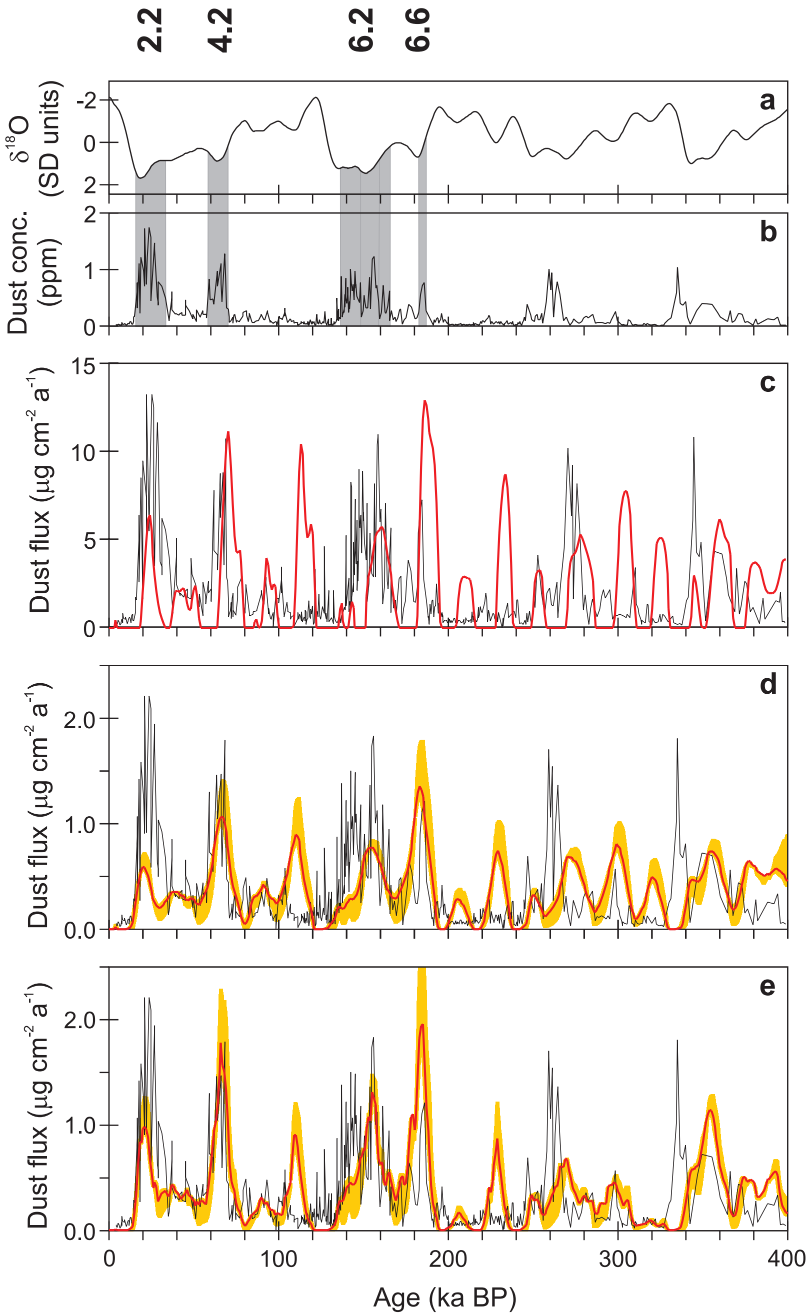

Figure 2. Observed and modeled relationship between dust and climate. (a) The SPECMAP $\delta^{18}O$ stack [*Imbrie et al.*, 1984] (which by convention is shown with the $\delta^{18}O$ scale reversed) with Marine Isotope Stages [*Bassinot et al.*, 1994] marked. (b) The Vostok dust concentration record [*Petit et al.*, 1999]. The apparent correspondence between prominent maxima in dust concentrations (Figure 2b) and periods of rapid increase in $\delta^{18}O$ (Figure 2a) during glacial times (loosely delineated by $\delta^{18}O$ greater than about 0.0 SD units) is shown highlighted (gray shading). In contrast to the rather close correlation over the earlier half of the record (0 to ~200 ka BP), there is no obvious analogous relationship between the two records over the older portion (~200 to 400 ka BP). (c) The Vostok dust depositional record (black line) reconstructed from dust concentrations [*Petit et al.*, 1999] and estimated snow accumulation history (J. B. Petit, personal communication, 2001) alongside the results of a simple (positive truncation) first-derivative model (red line). (d) Observations (black line) contrasted with the results of the initial source-only model (equation 3) (red line) with τ = 12.5 ka. Results from a sensitivity analysis with τ in the range 5–15 ka are marked in orange. (e) Observations (black line) contrasted with the results of the complete source + transport model (equation 4) (red line). Results from a sensitivity analysis with τ in the range 5–15 ka are marked in orange.

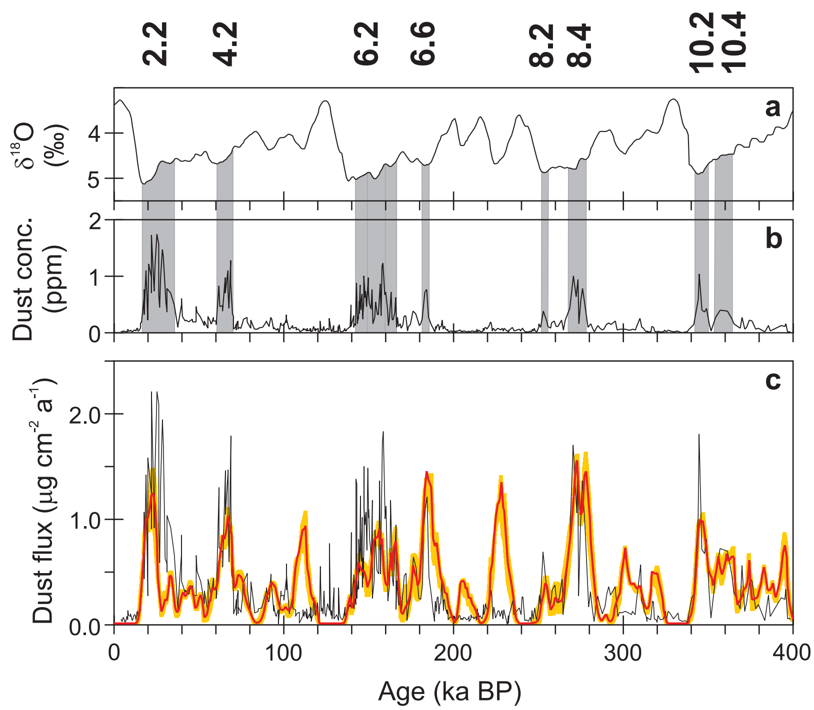

Figure 3. Observed and modeled relationship between dust and climate. (a) The benthic foraminiferal $\delta^{18}O$ record from V19-30 [*Shackleton*, 2000]. (b) The Vostok dust concentration record [*Petit et al.*, 1999], repotted on the same age scale as the marine $\delta^{18}O$ signal [*Shackleton*, 2000]. The apparent correspondence between prominent maxima in dust concentrations (Figure 3b) and periods of rapid increase in $\delta^{18}O$ (Figure 3a) is highlighted (gray shading). (c) Observations (black line) contrasted with dust fluxes (red line) predicted by the source + transport model (equation 4) with τ = 7.5 ka and taking the alternative $\delta^{18}O$ record as an input. Results from a sensitivity analysis with τ in the range 5–15 ka are marked in orange.