

Plio-Pleistocene African Climate

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Marine records of African climate variability document a shift toward more arid conditions after 2.8 million years ago (Ma), evidently resulting from remote forcing by cold North Atlantic sea-surface temperatures associated with the onset of Northern Hemisphere glacial cycles. African climate before 2.8 Ma was regulated by low-latitude insolation forcing of monsoonal climate due to Earth orbital precession. Major steps in the evolution of African hominids and other vertebrates are coincident with shifts to more arid, open conditions near 2.8 Ma, 1.7 Ma, and 1.0 Ma, suggesting that some Pliocene (Plio)-Pleistocene speciation events may have been climatically mediated.

Detailed records of African paleoclimate change during the Plio-Pleistocene are rare from terrestrial sequences, but deep-sea drilling of marine sedimentary sequences near the continent has recovered several continuous and well-dated records of wind-borne (eolian) dust variability, which serve as proxies of subtropical African climate change over the last ~5 million years. This interval is notably punctuated by the initial growth and subsequent expansion of high-latitude ice sheets near 2.8 Ma and 1.0 Ma, respectively (1–4). Although elements of high-latitude climate change are relatively well understood for this interval (4–7), the sensitivity of low-latitude climate to tropical and extratropical influences remains a central issue in paleoclimatology (7–10). In this article, I define Plio-Pleistocene changes in African climate using several detailed paleoclimate records from marine sediments off subtropical West and East Africa. The paleoclimate records and climate model experiments are used to explore high- and low-latitude influences on African terrestrial climate. These records are also linked to other records of African paleoenvironmental change and to the African fossil record of early hominid evolution to examine possible connections between climatic, ecologic, and faunal changes during the Plio-Pleistocene.

Subtropical African and Arabian soils supply prolific amounts of atmospheric dust to the marine environment. Sediment trap and mineralogic studies demonstrate that wind-borne detritus from these source areas comprises the dominant source of terrigenous sediment to the eastern equatorial Atlantic and the Arabian Sea (11, 12). Off West Africa, interannual variations in eolian dust concentrations are closely related to precipitation anomalies in subtropical African dust source areas (13). Dust storms are most frequent, and atmospheric dust loading is greatest, when the transition to prolonged drought reduces vegetational cov-

er, and dry soil surfaces are exposed to wind deflation (11, 14); other factors such as wind transport velocity and dust source area and trajectory changes are also important. Over geologic time scales, variations in subtropical African aridity are recorded stratigraphically as variable quantities of eolian detritus preserved in marine sediments (15).

Regional Climatic Setting and Paleoclimatic History

Today, subtropical African precipitation is highly seasonal and follows the annual cycle of the West African monsoon. During boreal summer, heating over the land surface draws moist maritime air from the equatorial Atlantic into western and central subtropical Africa (16) (Fig. 1A). East African summer rainfall is related to the westerly airstream of the African monsoon, but it is highly variable in part because of topographic rainshadow effects (17, 18). Strong southwesterly winds associated with the summer Asian monsoon develop along the Somali and Arabian coasts (Fig. 1A). Regional atmospheric circulation reverses during boreal winter, and dry, variable northeast trade winds blow over subtropical Africa, Arabia, and southeast Asia (Fig. 1B).

Well-defined dust plumes result from the strong seasonality of precipitation and transporting winds. A prominent summer dust plume off West Africa is carried by the African easterly jet, and its load is mainly derived from Saharan sources (Fig. 1A). The winter African dust plume originates from dry sub-Saharan and Sahelian soils which are carried by northeast trade winds (Fig. 1B). A summer dust plume off Arabia (Fig. 1A) originates from dry Mesopotamian, Arabian, and northeast African soils and is carried by northwest ("Shamal") and southwest Asian monsoon winds (11, 12).

Late Pleistocene African paleoclimate records indicate the dual but separate influences of high- and low-latitude processes (19). Enhanced African aridity during glacial maxima with evidence for cooler tem-

peratures has been documented in terrestrial and marine paleoclimate records (3, 19–26). There is also abundant evidence that the intensity of the African monsoon has been regulated by periodic [23- to 19-ky (thousand years)] variations in low-latitude insolation resulting from Earth orbital precession. Indicators of monsoon paleointensity, African lake levels, and tropical river outflow were greatest when perihelion (minimum Earth-sun distance) coincided with boreal summer, a configuration that increased summer season insolation which drives monsoonal circulation (19, 27–29).

Plio-Pleistocene changes in African climate must be evaluated within the context of significant changes in high-latitude climate which punctuate this interval. Marine oxygen isotopic records demonstrate that the onset of high-latitude glacial cycles developed gradually between 3.1 and 2.6 Ma (2, 3); glacial marine ice-rafted debris becomes abundant in polar ocean sediments near 2.7 to 2.8 Ma (4). Between 2.8 and 1 Ma, the high-latitude climate oscillated between moderate glacial and interglacial extremes at the 41-ky orbital obliquity (tilt) periodicity that regulates high-latitude seasonal insolation (2, 3, 5). Glacial climate extremes increased markedly after 1 Ma, and the dominant period of variation shifted to 100 ky (2, 3, 5, 30). High-latitude sea-surface temperatures (SSTs), ice-rafting, and deep circulation are strongly covariant with glacial ice volume fluctuations, indicating coherent high-latitude climate responses to orbital insolation forcing (2, 3, 5, 6).

Marine Eolian Records

Records of eolian dust variability were developed on the basis of eolian extraction analyses of marine sediment cores recovered from strategically located Ocean Drilling Program sites that lie beneath the dust plumes off West Africa (sites 659, 661, 662, 663, and 664) and East Africa and Arabia (sites 231 and 721/722; Fig. 1). Complete, composite sequences were constructed at sites 659, 661, 663, and 721/722 (3, 19, 31, 32). Site 659 off West Africa records variations in the summer West African dust supply (Fig. 1A), whereas sites 661, 662, 663, and 664 largely record dust supply variations from the winter West African plume (Fig. 1B). Sites 231 and 721/722 record past variations in the summer Arabian dust supply from northeast African, Arabian, and Mesopotamian sources (Fig.

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1A). Sites 231, 659, and 721/722 were drilled on bathymetric highs to minimize the effects of variable deep-ocean carbonate dissolution and hemipelagic sediment influences. Variable carbonate dissolution may have affected the terrigenous dust record in the deepest West African record [site 661; 4012 m (31)]. However, mass flux calculations of biogenic and terrigenous sediment variations at nearby sites 659 and 663 (Fig. 1) indicate that high terrigenous percent values at these sites reflect absolute increases in the mass flux of terrigenous eolian detritus rather than relative dilution effects due to variable carbonate production or dissolution (3, 19).

Terrigenous (eolian) dust records were developed at all sites with a variety of techniques. At sites 661 and 721/722, eolian percentages were estimated with continuous whole-core measurements of magnetic susceptibility, which is a rapid measure of magnetic particle concentration (31). Because eolian dust contains trace quantities of magnetic grains, susceptibility records at sites 661 and 721/722 were used to rapidly estimate eolian concentrations with analytic regressions (SE, ± 3.0 and ± 2.2 eolian percent, respectively). Terrigenous records at sites 662 and 663 were calculated as the residual from biogenic carbonate and opal analyses (19, 32). Terrigenous percent records at sites 231, 659, and 664 were calculated as the residual noncarbonate fraction of the sediment (3, 32).

Age control for each record was established by using the integrated oxygen isotopic, biostratigraphic, and magnetic polarity Plio-Pleistocene time scale (2, 33) and its application to site 659 (3). Average temporal resolution at all sites ranges between 1.2 ky and 3.5 ky. Site 659 is fully constrained throughout the Plio-Pleistocene by an oxygen isotopic stratigraphy (3). Sites 663 and 721/722 have detailed oxygen isotopic age control extending to 0.9 Ma and 1.1 Ma, respectively (19, 26). Additional age control at other sites was established with a combination of biostratigraphic and magne-

tostratigraphic datums. Detailed intercorrelations between the records were apparent, so small adjustments were permitted within the age control error limits to align the records and to correlate (tune) them to an orbital insolation composite (34). Site 721/722 is the longest complete dust record, extending to 7.3 Ma (35).

Site 231 sediments in the Gulf of Aden contain several ash horizons whose shards have been geochemically correlated to tephra layers within East African fluvio-lacustrine sequences (36). Disseminated shards of some of the same tephra layers have also been detected within the site 721/722 sediments (35). Age control of fossil-bearing sequences in East Africa is largely constrained by tephrostratigraphic correlations combined with precise radiometric dating. Extending these tephra correlations into marine sequences allows the fossil record of hominid and other vertebrate evolution to be placed within the context of very detailed records of regional paleoclimate variability.

Plio-Pleistocene Shifts in African Climate Variability

The marine eolian records exhibit marked shifts in concentration and variability centered near 2.8 Ma, 1.7 Ma, and 1.0 Ma (Fig. 2). A long-term increase in mean eolian concentration beginning near 2.8 Ma is apparent at sites 659, 662, 663, 664, and 721/722 (Fig. 2). Low-pass filtering of these records indicates that an inflection toward persistently higher baseline values occurs between 2.8 and 2.6 Ma. Mean late Pleistocene terrigenous concentrations at sites 659, 662, 663, and 664 are between two to five times the pre-2.8 Ma values (3, 32). The most pronounced increases in eolian values after 2.8 Ma occur at West African sites 663 and 664 (Fig. 2) which are most distal from Sahelian dust source areas. A short-lived but significant increase in mean dust concentrations is observed between 1.8 and 1.6 Ma at sites

659, 662, 664, and most significantly at site 721/722 (Fig. 2).

Dust layers deposited during times corresponding to high-latitude glaciations tend to be both thicker and have higher eolian concentrations than their interglacial counterparts, indicating that the increased dust abundances reflect real increases in the supply of wind-borne material (3, 19, 26). Glacial dust fluxes were calculated from sedimentation rate, bulk density, and eolian concentration data at sites 659, 663, and 721/722. During the latest Pleistocene, glacial interval dust fluxes were three to five times the interglacial fluxes at both West and East African sites (3, 19, 26). Like the oxygen isotopic records of global ice volume variability, the African dust flux records exhibit dominant 100-ky variability over the last 1 million years (3, 19, 26) (Fig. 2). Cross-spectral analyses between eolian dust and oxygen isotopic records (within the same cores) demonstrate that Pleistocene dust flux maxima are coincident with or slightly lead glacial maxima (3, 19, 26). Latest Pleistocene lacustrine records from subtropical Africa demonstrate that the development of arid conditions was essentially synchronous with high-latitude cold events (22, 25).

Shifts in the dominant period of eolian variability are detected near 2.8 ± 0.2 Ma, 1.7 ± 0.1 Ma, and 1.0 ± 0.2 Ma. The timing and magnitude of these shifts was quantified at each site with evolutive power spectral analysis which determines dominant periodicities of variation for a series of discrete (for example, 800-ky length; Fig. 2) and overlapping (300-ky time step) power spectra (37). Precise age-estimates of variance transitions (± 0.1 Ma) were obtained at each site by using smaller spectral data windows and shorter time step values. The site 721/722 evolutive power spectra are shown for example in Fig. 2. The changes in eolian variance described below were detected at all other sites with comparable age control (for example, sites 659, 661, 662, and 663).

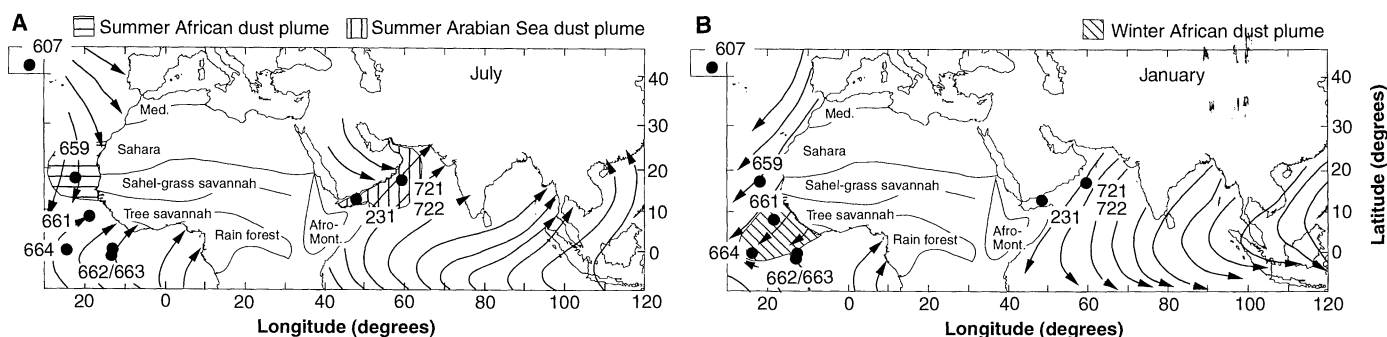


Fig. 1. West and East African Ocean Drilling Program site locations showing boreal summer (A) and winter (B) surface wind and eolian dust trajectories (11, 16). Med., Mediterranean; Afro-Mont., Afro-Montaine vegetation zones.

Before 2.8 Ma, the spectra of the dust records indicate that subtropical African climate varied primarily at precessional (23- to 19-ky) periodicities (Fig. 2); this mode of variability extended at least into the mid-Miocene (~12 Ma) as indicated by the longest record at site 721/722. After 2.8 Ma there was a marked increase in 41-ky variance which persisted until ~1.0 Ma. Large-amplitude 41-ky eolian cycles are detected at sites 659, 661, 664, and 721/722 only after 1.6 to 1.7 Ma. At site 721/722 this shift is accompanied by a marked increase in eolian concentrations (Fig. 2). Near 1.0 Ma, the eolian cycles shifted from a dominant 41-ky period to a larger duration and amplitude 100-ky period. Detailed evolutive spectral analyses demonstrate that the

variance transition from 23- to 19-ky to 41-ky eolian cycles occurred between 3.0 and 2.7 Ma at site 659, between 3.1 and 2.7 Ma at site 661, between 3.0 and 2.8 Ma at site 662, and between 3.1 and 2.7 Ma at site 721/722. Strong precessional variance persisted at site 659 until ~1.6 Ma (3). The increase in 100-ky variance occurred between 1.1 and 0.7 Ma at site 659, 1.1 and 0.7 Ma at site 661, and 1.2 and 0.9 Ma at site 721/722.

The shifts in African climate variability recorded in the dust records are coeval with changes in high-latitude climate. The 2.8-Ma increase in 41-ky eolian variance and concentration coincides with the onset of bipolar glaciation and the subsequent development of 41-ky glacial cycles between

3.0 and 2.6 Ma (2–4, 31). The dominance of 41-ky eolian variance after 1.6 Ma parallels the development of enhanced 41-ky high-latitude glacial climate cycles (2, 5). The 1.0-Ma increase in 100-ky eolian variance coincides with the marked increase in glacial climate amplitude after 1.2 to 0.9 Ma and the development of 100-ky climate cycles (2, 4, 26, 31).

African Climate and North Atlantic Sea-Surface Temperatures

Modern occurrences of Sahelian drought have been related to cold SST anomalies in the North Atlantic and relatively warm SST anomalies in the South and Equatorial Atlantic (7, 9, 19, 38, 39). This configu-

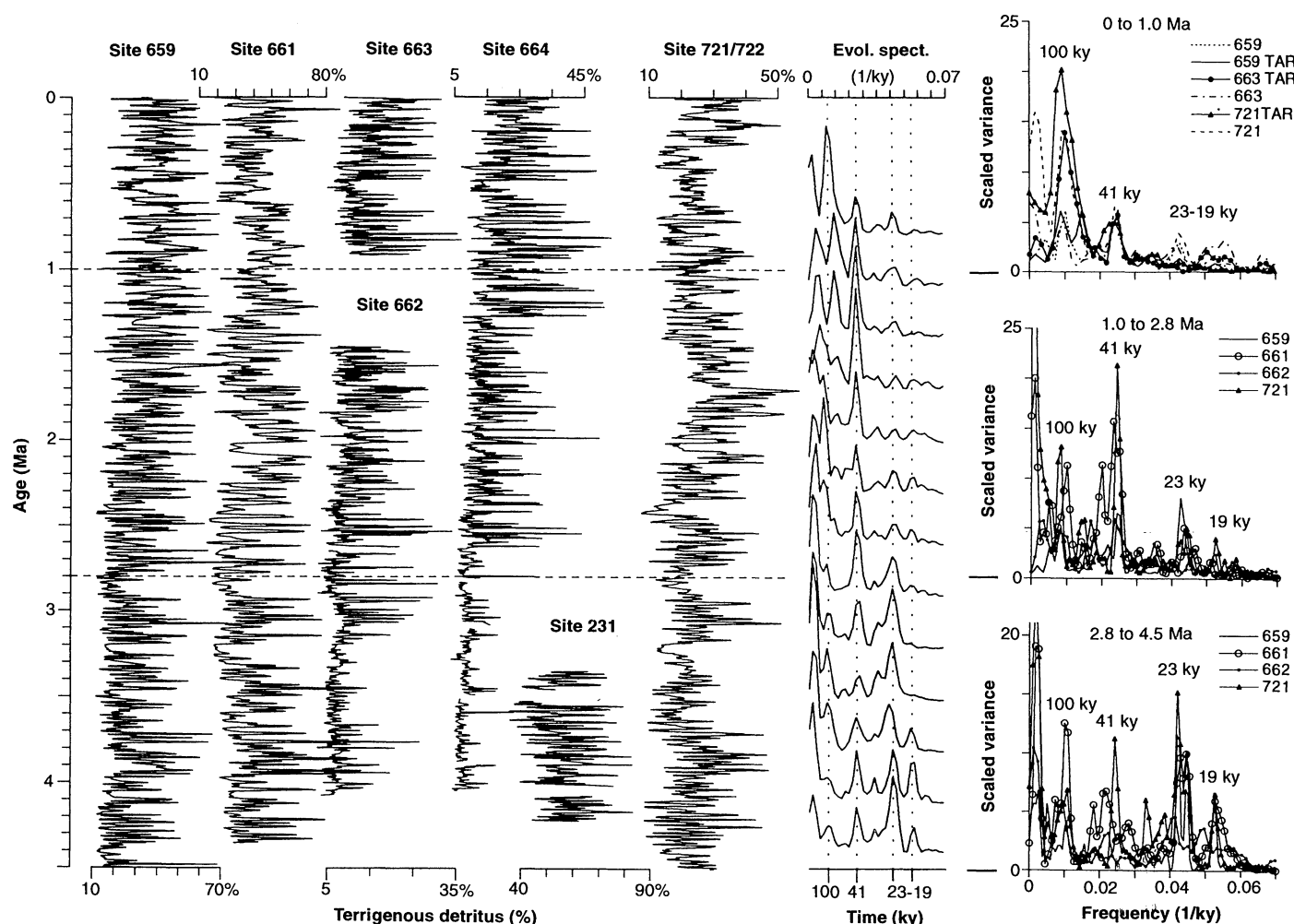


Fig. 2. Percent of terrigenous (eolian) detritus in cores from West African sites 659, 661, 662, 663, and 664, and East African and Arabian sites 231 and 721/722 (3, 19, 31, 32, 35). Age control was established with the integrated oxygen isotopic, magnetic polarity, and biostratigraphic time scale developed by Shackleton and others (2, 3, 33); see text for data and time scale information. Discrete (far right) and evolutive (right, site 721/722 record example) power spectral analyses (Evol. spect.) with the Blackman-Tukey method (37) (linear detrend, no prewhitening, one-third lag, common scaled variance) were used to summarize modes of African climate variability over time. Pronounced shifts in African climate variability occur near 2.8 ± 0.2 Ma, 1.7 ± 0.1 Ma, and 1.0 ± 0.2 Ma. African climate varied primarily at periodicities corresponding to

orbital precession (23 and 19 ky) until ~2.8 Ma. After 2.8 Ma, the dominant mode of variation shifted to the 41-ky periodicity, coeval with the onset of 41-ky high-latitude glacial climate cycles. The predominance of 41-ky eolian variance after 1.7 Ma parallels the development of larger amplitude 41-ky high-latitude glacial ice volume cycles. After 1 Ma, eolian variability shifted to a dominant 100-ky periodicity, coincident with the development of larger amplitude 100-ky glacial climate cycles. Strong 100-ky eolian variance after 1 Ma is particularly apparent in the power spectra of eolian mass flux records (sites 659, 663, 721; upper right). Cross spectral analyses of both West and East African records indicate that maximum dust supply coincides with, or slightly precedes glacial maxima (3, 19, 26).

ration inhibits the penetration of the summer African monsoon rainfall and tends to enhance winter trade wind velocities (40). The El Niño–Southern Oscillation (ENSO) also significantly affects African precipitation (18, 41). In accord with historical observations, African paleoclimate studies demonstrate that subtropical West, Central, and East Africa were more arid when North Atlantic SSTs were cold during Pleistocene glacial intervals (19, 21–22, 24, 42).

At West African site 663 (Fig. 1), a 900-ky record of eolian dust and savannah grass phytolith (windblown opaline grass cuticles) concentrations (19) covary with a fossil-faunal record of North Atlantic SST variability from site 607 [41°N (5)]. All records are dominated by strong and coherent 100-ky and 41-ky variations (19); dust and phytolith concentrations were highest when North Atlantic SSTs were coldest (up to -12°C cooler). Strong covariance between the terrigenous and phytolith records at site 663 suggests that the cold glacial North Atlantic SSTs affected both African dust generation and transport and regional vegetation. Marine pollen records have documented that West African vegetation zones migrated southward during Plio-Pleistocene glacial stages (24).

Climate Model Experiments

The National Aeronautics and Space Administration–Goddard Institute for Space Studies (NASA-GISS) atmospheric general circulation model has been used to examine the sensitivity of subtropical African climate to cold North Atlantic SSTs (7, 9, 39). The model solves the equations for conservation of mass, energy, momentum, and moisture for nine atmospheric layers for an 8° by 10°

grid resolution (7). It calculates cloud cover, snow cover, soil moisture, and full radiative processes with a diurnal and seasonal cycle; SSTs are prescribed and noninteractive in all runs. The control model run (modern climatology) was reconfigured with cold, glacial SSTs (43) in the North Atlantic basin north of 25°N (7). Climate anomalies resulting from these cold SSTs are expressed in terms of their departures from the control model climatology (Fig 3, A and B).

Cold North Atlantic SSTs cause subtropical West and Central Africa to become annually cooler and drier. During the winter months (December, January, February), cold North Atlantic SSTs strengthen the subtropical Atlantic high-pressure cell and thus greatly enhance northeast trade winds over West Africa (Fig. 3A). Winter trade wind speeds off West Africa increased 180% relative to control values (Fig. 3). Averaged over the full subtropical African domain (Fig. 4), the v -component (north-south) of the trade winds increased 65% over control values. This circulation advects cooler (1° to 4°C) and drier European continental air over subtropical Africa. During the summer months (June, July, August), anticyclonic circulation associated with the cold North Atlantic SSTs opposes (weakens) the moist westerly (cyclonic) monsoonal inflow into subtropical Africa up to 50% relative to control values, as evidenced by the easterly difference vectors off West Africa (Fig. 3B) and the reduced summer precipitation values (Fig. 4). Surface temperatures are locally depressed by 1° to 6°C (Fig. 3), but the broader regional cooling is only 1.3°C annually (Fig. 4). Annual rainfall in subtropical Africa was reduced by 30% (lower by -0.35 mm day^{-1} relative to the control mean value of 1.19

mm day^{-1}), with the largest reductions occurring in summer (Fig. 4).

Tropical SSTs were unchanged in these experiments. When tropical SSTs are reduced by the 5°C indicated by glacial-age tropical coral, pollen, and mountain snow-line studies (7, 10, 20, 21, 44), subtropical and tropical Africa become dramatically cooler and drier (7). Cooler tropical SSTs both sensibly cool the continent and greatly reduce evaporative moisture fluxes feeding the monsoonal rains (as a result of the nonlinear Clausius–Clapeyron relation linking SST and saturation vapor pressure). Cooler glacial tropical SSTs may reflect changes in the tropical troposphere [increased low-level cloud cover (10)], or a more fundamental change in ocean heat transport from the tropic oceans (45).

African Climate Variability Before 2.8 Ma

I measured variations in eolian detritus, biogenic opal (marine diatom and radiolarian abundances), and organic carbon in the site 721/722 precessional cycles between 5.9 and 5.4 Ma to examine the paleoclimatic origin of the precessional climate cycles before 2.8 Ma. In the modern Arabian Sea, surface productivity and eolian dust supply are coupled to the summer Asian monsoon circulation; dust-laden southwest winds establish intense coastal upwelling and high productivity off the Arabian and Omani margins (16, 28). The eolian dust, opal, and organic carbon records vary coherently and in-phase with each other and closely follow the calculated July (30°N) orbital insolation forcing (Fig. 5).

These results suggest that the terrestrial climate and marine upwelling signatures of

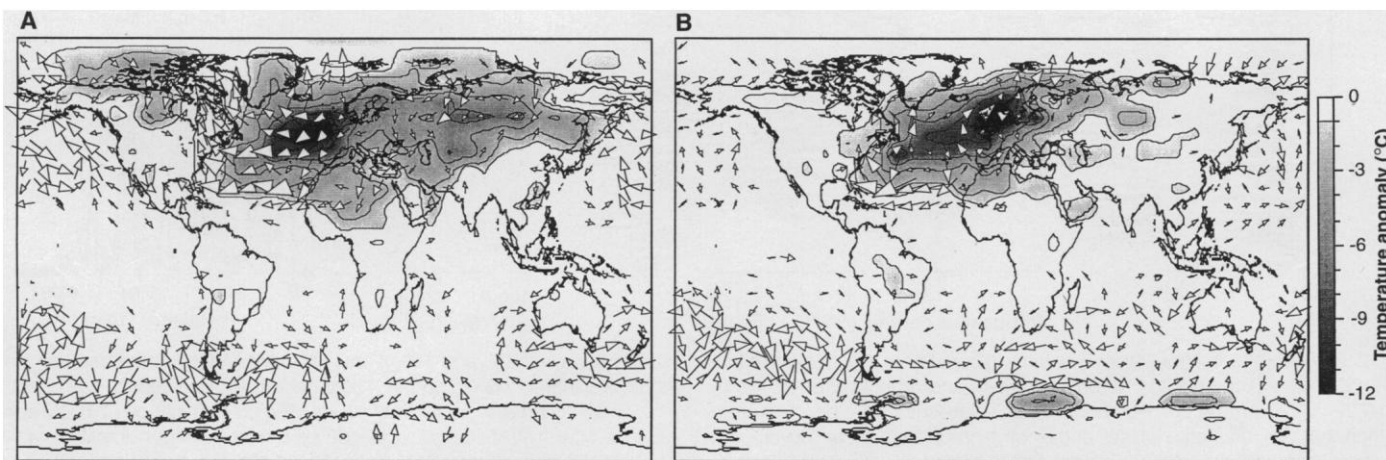


Fig. 3. Surface temperature and wind anomalies for boreal winter (December, January, February) (A) and summer (June, July, August) (B) due to cold CLIMAP SSTs prescribed for the North Atlantic basin north of 25°N with the 8° by 10° NASA-GISS climate model (7, 9). (A) shows that colder SSTs in the North Atlantic enhance the subtropical high pressure cell which increases northeast trade winds over West Africa and advects cooler (1° to 4°C) and

drier European air over the continent. (B) shows that during the summer months this high pressure system opposes the westerly monsoonal inflow of moist maritime air into West and Central Africa (westward-pointing difference vectors) and advects cooler, drier air over western and central subtropical Africa. Temperature anomaly contours are -10° , -8° , -6° , -4° , -2° , -1° , and $+1^{\circ}\text{C}$; plotted wind difference vectors are 0.5 to 4.0 m/s.

the Asian monsoon were tightly coupled to direct precessional insolation forcing before 2.8 Ma. More generally, the recurrence of periodically wet and dry climates before 2.8 Ma was apparently regulated by orbital changes in direct low-latitude radiation forcing of monsoonal climate. Orbital precession affects the seasonal receipt of low-latitude summer insolation which, in turn, drives the monsoon. Prell and Kutzbach (28) have shown that monsoonal precipitation and surface winds increased by 38 and 75%, respectively, in response to a 19% increase in summer insolation due to orbital precession.

Plio-Pleistocene African Climate Change

African climate evidently responded to direct precessional insolation forcing of monsoonal climate before 2.8 Ma, when high-latitude ice sheets were small and relatively invariant. After 2.8 Ma, when ice sheets grew such that large glacial cycles were sustained, African climate became sensitive to remote changes in high-latitude climate. The data and model results suggest that African climate became periodically cooler and drier after 2.8 Ma as a result of dynamical effects related to the development of cold glacial North Atlantic SSTs; this effect was further amplified after 1 Ma following the increase in the duration and magnitude of high-latitude glacial cycles. Paleoclimate evidence demonstrates, however, that precessional forcing of monsoonal climate persisted throughout the entire Plio-Pleistocene (19, 26–29, 46). The high-latitude influence is thus viewed to be a superimposed but primary factor affecting African terrestrial climate after 2.8 Ma (19, 35).

East African vegetation shifted from closed canopy to open savannah vegetation starting in the mid-Pliocene, marking a progression toward reduced and seasonally contrasted precipitation. Late Miocene to mid-

Pliocene (8 to 3 Ma) fossil floral and isotopic evidence indicates that subtropical West and East Africa were considerably warmer and more humid than at present, supporting annually wet lowland rain forest vegetation in regions that today support seasonally dry savannah grasses and shrubs (20, 47–50). The expansion of arid-adapted flora and fauna near 2.8 Ma may have occurred rapidly (50–53) or more gradually (49). Lessons from latest Pleistocene paleoclimate records demonstrate that African climate and vegetation responded swiftly to abrupt changes in high-latitude temperatures (21, 22, 25, 42), so a similar sensitivity to the onset of high-latitude glacial conditions near 2.8 Ma may be expected. Indeed, a detailed Plio-Pleistocene pollen record off West Africa [site 658; 21°N, 19°W; see (50)] indicates that humid (rain forest-affinity) taxa were replaced by arid-adapted (*Artemisia* and *Ephedra*) taxa between 3.2 and 2.6 Ma; trade wind transport of Mediterranean taxa increased at this time as well. Constructional uplift of East African volcanic provinces may have contributed also to the vegetation changes observed at some localities (47, 48).

On the Paleoenvironment of Hominid Evolution

Some of the major events in early hominid evolution appear to be coeval with these African climate changes, supporting previous assertions that certain junctures in human evolution may have been climatically mediated (52–57). The fossil record is still too fragmentary to establish precise relations with paleoclimate records, taphonomic biases are always operating, and coincidence cannot in itself imply causality. Accepting these caveats, the marine eolian records provide an opportunity to examine the available African fossil record of hominid and other vertebrate evolution within the context of regional paleoenvironmental change.

The most significant development in early hominid evolution occurs between 3.0 and 2.5 Ma when at least two distinct lineages emerge from a single bipedal ancestral line (Fig. 6). Earliest members of the “robust” australopithecine lineage first occur in the fossil record near 2.7 Ma (57–59) (Fig. 6) and are distinguished by apparently unique masticatory adaptations (54, 56, 57). A second lineage, represented by the earliest members of our genus *Homo*, first occurs in East African sediments dated near 1.9 Ma (58, 59); recent but fragmentary discoveries extend this range to 2.5 Ma (60) (Fig. 6). Earliest fossils of the *Homo* clade are characterized by larger absolute cranial volumes (61). The earliest known stone tools (crude choppers and scrapers composing the Oldowan complex) occur near 2.4 to 2.6 Ma (62). The synchronous existence of two distinct hominid lineages has been interpreted to reflect separate adaptations to a more arid, varied environment (56, 57). Fossil African bovid and rodent assemblages indicate shifts toward arid-adapted species between 2.7 and 2.5 Ma (52, 53, 55).

By 1.6 Ma, *Homo habilis* became extinct; its immediate successor, and our direct ancestor, *H. erectus*, first occurs in the fossil record near 1.8 Ma (58, 59, 61). *Homo erectus* may have migrated to southeast Asia as early as 1.8 to 1.6 Ma (63), although its taxonomic affinity with African *H. erectus*

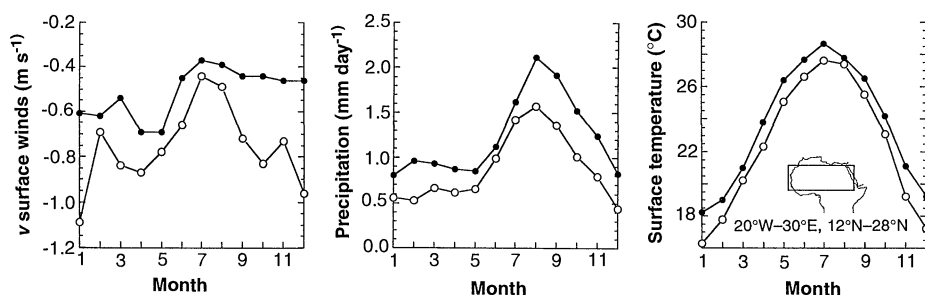


Fig. 4. Monthly surface temperature, precipitation, and v-component (north-south) surface wind data for the NASA-GISS climate model control run (filled circles) and the cold North Atlantic SST model runs (open circles). Data were calculated as monthly averages for the region between 20°W and 30°E, and 12°N and 28°N (see inset). These model results illustrate that cold North Atlantic SSTs produce significant annual cooling and drying of subtropical Africa and that the responses have seasonal signatures. Cool North Atlantic SSTs enhance winter trade wind circulation, reduce summer monsoonal rainfall, and cool the region most dramatically in the winter months.

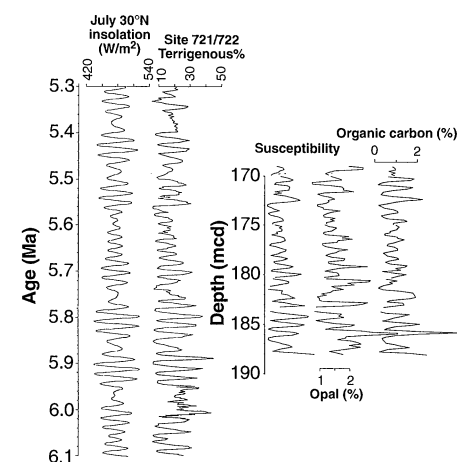


Fig. 5. Paleoclimatic origin of the precessional (23- to 19-ky) climate cycles at Arabian Sea site 721/722. In the modern Arabian Sea, surface productivity and eolian dust supply are greatest during the summer Asian monsoon when dust-laden southwest winds establish intense coastal upwelling off the Arabian and Omani margins (12, 16). Sedimentary indices of Arabian and East African dust transport (terrigenous percent) and Arabian Sea surface productivity (biogenic opal and organic carbon) are coherent and in-phase with each other and with a July insolation time series calculated for 30°N (34). These results indicate that the site 721/722 precessional cycles reflect direct orbital insolation forcing of the Asian monsoon intensity. Mcd, meters composite depth.

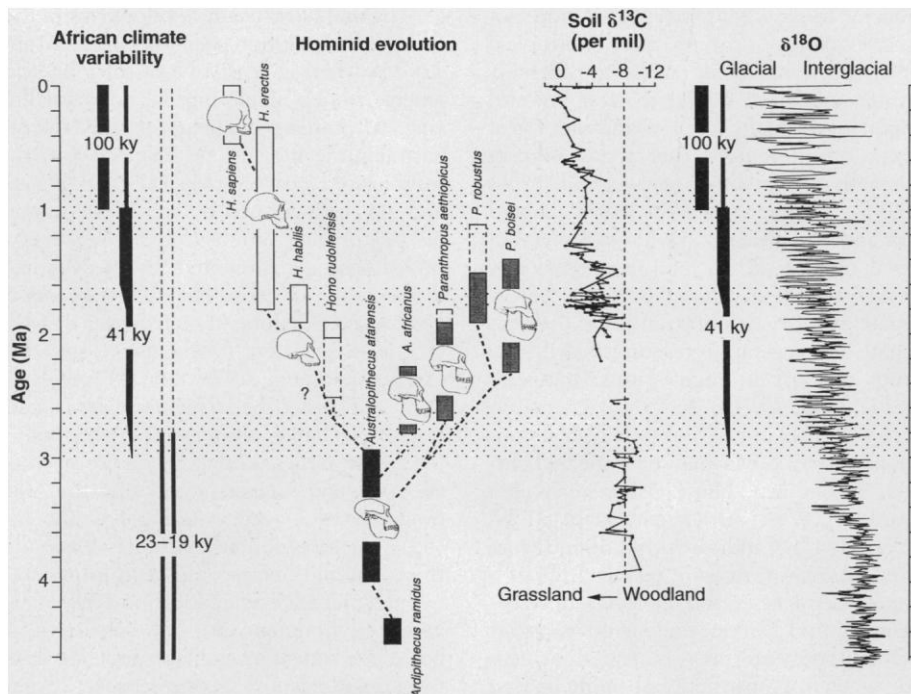


Fig. 6. Temporal relations between changes in African climate, hominid evolution, East African vegetation, and glacial ice volume [composite record based on data from (2, 5, 33)]. Step-like increases in African aridity and shifts in variability near 2.8, 1.7, and 1.0 Ma are evidently coeval with significant events in early hominid and other vertebrate evolution in East Africa. At least two distinct hominid lineages (*Paranthropus* and *Homo* genera) emerge from a single ancestral line (*Australopithecus afarensis*) between 3.0 and 2.5 Ma (58, 59, 61). Rapid expansion of arid-adapted biota near 2.5 to 2.6 Ma is indicated by fossil African vegetation and fossil bovid and micromammal evidence (52, 53, 55). Increased regional aridity near 1.8 to 1.6 Ma indicated by the marine records (Fig. 2) is supported by soil carbonate stable isotopic evidence ($\delta^{13}\text{C}$ values) for expanded savannah vegetation in East Africa (49). By 1.6 Ma, *Homo habilis* had become extinct and was succeeded by *H. erectus*, our direct ancestor. "Robust" australopithecines (*Paranthropus*) had become extinct by 1.4 Ma (57–59), and by 1 Ma *H. erectus* had broadly expanded its geographic range and occupied sites in North Africa, Europe, and western Asia (65). The coincidence between these African climatic and biotic shifts suggests that development of periodically cooler and drier African conditions near 2.8, 1.7, and 1.0 Ma may have modified the African biotic landscape in favor of arid-adapted species.

is debated. Near 1.7 Ma, East African bovid assemblages shift toward further absolute increases in the abundance of arid-adapted species (52, 55). The earliest occurrences of the more sophisticated Acheulean tool kit (bifacial handaxes) occur near 1.4 Ma (64). Enhanced African aridity near 1.8 to 1.6 Ma at site 721/722 is supported by soil carbonate stable isotopic evidence for broadly expanded savannah vegetation in East Africa (49) (Fig. 6). The "robust" australopithecine lineage became extinct near 1.4 Ma (57, 59), although for taphonomic reasons this datum may be considerably younger (57, 59). By 1 Ma *H. erectus* had broadly expanded its geographic range and occupied sites in North Africa, Europe, and western Asia (65). The fossil record of African bovidae documents a final phase of increased arid-adapted species composition near 1 Ma (52, 55).

The marine paleoclimate records demonstrate that Plio-Pleistocene African climate change is most accurately characterized as a continuum of alternating wet and

dry conditions with significant punctuations in eolian variability and concentration occurring near 2.8, 1.7, and 1.0 Ma. The development of periodically cooler and drier African conditions after 2.8 Ma, and their subsequent intensification after 1.7 and 1.0 Ma, may have established discrete opportunities for ecologic fragmentation and genetic isolation leading to the eventual rise of arid-adapted species.

REFERENCES AND NOTES

- N. J. Shackleton et al., *Nature* **307**, 620 (1984).
- N. J. Shackleton, A. Berger, W. R. Peltier, *Trans. R. Soc. Edinburgh Earth Sci.* **81**, 251 (1990).
- R. Tiedemann, M. Samthein, N. J. Shackleton, *Paleoceanography* **9**, 619 (1994).
- E. Jansen and J. Sjöholm, *Nature* **349**, 600 (1991).
- W. F. Ruddiman, M. E. Raymo, D. G. Martinson, B. M. Clement, J. Backman, *Paleoceanography* **4**, 353 (1989).
- M. E. Raymo, W. F. Ruddiman, N. J. Shackleton, D. W. Oppo, *Earth Planet. Sci. Lett.* **97**, 353 (1990).
- D. Rind and D. Peteet, *Quat. Res.* **24**, 1 (1985).
- W. S. Broecker and G. H. Denton, *Geochim. Cosmochim. Acta* **53**, 2465 (1989).
- P. B. deMenocal and D. Rind, *J. Geophys. Res.* **98**, 7265 (1993).
- T. P. Guilderson, R. G. Fairbanks, J. L. Rubenstein, *Science* **263**, 663 (1994).
- K. Pye, *Eolian Dust and Dust Deposits* (Academic Press, New York, 1987).
- F. Sirocko and M. Samthein, in *Paleoclimatology and Paleometeorology: Modern and Past Patterns of Global Atmospheric Transport*, M. Leinen and M. Samthein, Eds., NATO ASI Ser. **1989**, 401 (1989); F. Sirocko, M. Samthein, H. Lange, H. Erlenkeuser, *Quat. Res.* **36**, 72 (1991); R. R. Nair et al., *Nature* **338**, 749 (1989).
- J. M. Prospero and R. T. Nees, *Science* **196**, 1196 (1977); *Nature* **320**, 735 (1986).
- N. J. Middleton, *Nature* **316**, 431 (1985).
- D. Rea, *Rev. Geophys.* **32**, 159 (1994).
- S. Hastenrath, *Climate and Circulation of the Tropics* (Reidel, Boston, 1985).
- T. D. Davies, C. E. Vincent, A. K. C. Beresford, *J. Climatol.* **5**, 17 (1985).
- S. E. Nicholson, in *The Limnology, Climatology, and Paleoclimatology of the East African Lakes*, T. Johnson and E. Odada, Eds. (Gordon and Breach, Newark, NJ, in press).
- P. B. deMenocal, W. F. Ruddiman, E. M. Pokras, *Paleoceanography* **8**, 209 (1993).
- A. C. Hamilton, *Environmental History of East Africa* (Academic Press, New York, 1982).
- R. Bonnefille, J. C. Roeland, J. Guiot, *Nature* **345**, 347 (1990).
- F. Gasse, R. Téhé, A. Durand, E. Gibert, J. C. Fontes, *ibid.* **346**, 141 (1990); F. A. Street-Perrott and R. A. Perrott, *ibid.* **343**, 607 (1990).
- M. Samthein, G. Tetzlaff, B. Koopman, K. Wolter, U. Pflaumann, *ibid.* **293**, 193 (1981).
- L. Dupont and H. Hoogheemstra, *Acta Bot. Neerl.* **38**, 405 (1989).
- F. Gasse, V. Lédée, M. Massault, J. C. Fontes, *Nature* **342**, 57 (1989).
- S. Clemens and W. J. Prell, *Proc. Ocean Drill. Program Sci. Res.* **117**, 365 (1991).
- M. Rossignol-Strick, *Nature* **303**, 46 (1983); F. A. Street-Perrott and S. A. Harrison, in *Climate Processes and Climate Sensitivity*, J. E. Hansen and T. Takahashi, Eds. (American Geophysical Union, Washington, DC, 1984), pp. 118–129; E. M. Pokras and A. C. Mix, *Quat. Res.* **24**, 137 (1985).
- W. L. Prell and J. E. Kutzbach, *J. Geophys. Res.* **92**, 8411 (1987).
- A. McIntyre, W. F. Ruddiman, K. Karlin, A. C. Mix, *Paleoceanography* **4**, 19 (1989); B. Molino and A. McIntyre, *Science* **249**, 766 (1990).
- J. Imbrie et al., *Paleoceanography* **8**, 699 (1993).
- J. Bloemendal and P. B. deMenocal, *Nature* **342**, 897 (1989).
- W. F. Ruddiman and T. Janecek, *Proc. Ocean Drill. Program Sci. Res.* **108**, 211 (1989).
- N. J. Shackleton, S. Crowhurst, T. Hagelberg, N. J. Pisias, D. A. Schneider, *ibid.*, in press.
- A. Berger and M. F. Loutre, *Quat. Sci. Rev.* **10**, 297 (1991).
- P. B. deMenocal and J. Bloemendal, in *Paleoclimate and Evolution with Emphasis on Human Origins*, E. Vrba, G. Denton, L. Burckle, T. Partridge, Eds. (Yale Univ. Press, New Haven, CT, in press).
- F. H. Brown, A. M. Sama-Wojcicki, C. E. Meyer, B. Haileab, *Quat. Int.* **13/14**, 55 (1992); A. M. Sama-Wojcicki, C. E. Meyer, P. H. Roth, F. H. Brown, *Nature* **313**, 306 (1985).
- R. B. Blackman and J. W. Tukey, *The Measurement of Power Spectra from the Point of Communications Engineering* (Dover, New York, 1958), pp. 1–258.
- P. J. Lamb, *Mon. Weather Rev.* **106**, 482 (1978); C. Folland, T. Palmer, D. Parker, *Nature* **320**, 602 (1986).
- L. Druyan, *Int. J. Climatol.* **9**, 77 (1989).
- R. E. Newell and J. W. Kidson, *J. Climatol.* **4**, 27 (1984).
- M. Cane, G. Eshel, R. W. Buckland, *Nature* **370**, 204 (1994).
- F. Gasse and E. Van Campo, *Earth Planet. Sci. Lett.* **126**, 435 (1994).
- CLIMAP Project Members, *Geol. Soc. Am. Map Chart Ser.* **36** (1981).
- P. Webster and N. Streten, *Quat. Res.* **10**, 279 (1978).
- R. Webb, S. Lehmann, D. Rind, R. J. Healey, *Eos* **75** (no. 44), 381 (1994).

46. F. J. Hilgen and C. G. Langereis, *Terra Nova* **1**, 409 (1989).
47. K. Yemane, R. Bonnefille, H. Faure, *Nature* **318**, 653 (1985).
48. R. Bonnefille, A. Vincens, G. Buchet, *Palaeogeogr. Palaeoclimatol. Palaeoecol.* **60**, 249 (1987); G. WoldeGabriel *et al.*, *Nature* **371**, 330 (1994).
49. T. E. Cerling, *ibid.* **97**, 241 (1992).
50. S. Leroy and L. Dupont, *ibid.* **109**, 295 (1994).
51. R. Bonnefille, *Nature* **303**, 487 (1983).
52. E. S. Vrba, G. H. Denton, M. L. Prentice, *Ossa* **14**, 127 (1989).
53. H. B. Wesselman, *S. Afr. J. Sci.* **81**, 260 (1985).
54. C. K. Brain, *Annex Transv. Geol. Soc. S. Afr. J. Sci.* **84**, 1 (1981).
55. E. S. Vrba, in *Paleoclimate and Evolution with Emphasis on Human Origins*, E. Vrba, G. Denton, L. Burckle, T. Partridge, Eds. (Yale Univ. Press, New Haven, CT, in press).
56. F. E. Grine, *S. Afr. J. Sci.* **82**, 87 (1986).
57. R. G. Klein, in *Evolutionary History of the "Robust" Australopithecines*, F. E. Grine, Ed. (de Gruyter, New York, 1988).
58. C. S. Feibel, F. H. Brown, I. McDougall, *Am. J. Phys. Anthropol.* **78**, 595 (1989).
59. W. Kimbel, in (55).
60. F. Schrenk, T. G. Bromage, C. G. Betzler, U. Ring, Y. M. Juwayeyi, *Nature* **365**, 833 (1993); A. Hill, S. Ward, A. Dieno, G. Curtis, R. Drake, *ibid.* **355**, 719 (1992).
61. B. Wood, *ibid.* **355**, 783 (1992).
62. J. W. K. Harris, in *African Archaeological Review*, N. David, Ed. (Cambridge Univ. Press, Cambridge, 1983), p. 3.
63. C. C. Swisher III *et al.*, *Science* **263**, 1118 (1994).
64. B. Aslaw *et al.*, *Nature* **362**, 732 (1992).
65. J. D. Clark, in *Human Ecology in Savanna Environments*, D. R. Harris, Ed. (Academic Press, London, 1980), pp. 41–71.
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RESEARCH ARTICLE

Sulfite Reductase Structure at 1.6 Å: Evolution and Catalysis for Reduction of Inorganic Anions

Brian R. Crane, Lewis M. Siegel, Elizabeth D. Getzoff*

Fundamental chemical transformations for biogeochemical cycling of sulfur and nitrogen are catalyzed by sulfite and nitrite reductases. The crystallographic structure of *Escherichia coli* sulfite reductase hemoprotein (SiRHP), which catalyzes the concerted six-electron reductions of sulfite to sulfide and nitrite to ammonia, was solved with multiwavelength anomalous diffraction (MAD) of the native siroheme and Fe_4S_4 cluster cofactors, multiple isomorphous replacement, and selenomethionine sequence markers. Twofold symmetry within the 64-kilodalton polypeptide generates a distinctive three-domain α/β fold that controls cofactor assembly and reactivity. Homology regions conserved between the symmetry-related halves of SiRHP and among other sulfite and nitrite reductases revealed key residues for stability and function, and identified a sulfite or nitrite reductase repeat (SNIIR) common to a redox-enzyme superfamily. The saddle-shaped siroheme shares a cysteine thiolate ligand with the Fe_4S_4 cluster and ligates an unexpected phosphate anion. In the substrate complex, sulfite displaces phosphate and binds to siroheme iron through sulfur. An extensive hydrogen-bonding network of positive side chains, water molecules, and siroheme carboxylates activates S-O bonds for reductive cleavage.

Sulfite and nitrite reductases (SiRs and NiRs) are key to both biosynthetic assimilation of sulfur and nitrogen and dissimilation of oxidized anions for energy transduction (1). Found throughout the three major kingdoms of living organisms (Archaea, Bacteria, and Eucarya), SiRs and most NiRs employ a siroheme (reduced porphyrin of the isobacteriochlorin class) (2) that is exchange-coupled with an iron-sulfur cluster to perform the remarkable reduction of a single atomic center by six electrons (3).

Assimilatory SiRs and NiRs in bacteria,

fungi, algae, and plants provide the reduced sulfur (oxidation state -2) and nitrogen (oxidation state -3) necessary for incorporation into biomolecules required by themselves and other higher organisms (1, 4). SiR generates sulfide from sulfite for subsequent cysteine biosynthesis in the terminal step of the 3'-phosphoadenylyl sulfate (PAPS) pathway (4). Assimilatory reduction of nitrate to ammonia proceeds by initial two-electron reduction to nitrite and then direct six-electron reduction to ammonia by an NiR that contains a siroheme and an iron-sulfur cluster (1). In contrast, dissimilatory denitrification transforms nitrate to nitrite, nitric oxide, nitrous oxide, and finally dinitrogen through sequential one- and two-electron reductions (1). The assimilatory chemistry is responsible for re-

ducing more than 10^4 megatons of NO_3^- a year, thus producing 100 times more NH_3 than nitrogen fixation by nitrogenase (5).

During dissimilatory sulfite reduction in sulfate-reducing eubacteria and some thermophilic archaeobacteria, sulfite can act as a terminal electron acceptor during anaerobic respiration (1, 4). Excess sulfide released to the environment contributes to biogeochemical sulfur cycling and causes corrosion and contamination problems for the oil and sewage treatment industries (6). Dissimilatory SiRs have siroheme and iron-sulfur clusters, are multimeric with varying subunit composition, and are differentiated from assimilatory sulfite reductases by their propensity to release trithionate ($\text{S}_3\text{O}_6^{2-}$) and thiosulfate ($\text{S}_2\text{O}_3^{2-}$) byproducts (1).

The *E. coli* assimilatory SiR (E.C. 1.8.1.2) is an oligomer of eight 66-kD flavoprotein (SiRFP) and four 64-kD hemoprotein (SiRHP) subunits (7, 8). In vivo, SiRFP transfers electrons from reduced nicotinamide adenine dinucleotide phosphate (NADPH) to SiRHP (9). Each SiRFP has one flavin adenine dinucleotide (FAD) and one flavin mononucleotide (FMN) binding site, yet the SiRFP octamer binds only four FAD and four FMN cofactors (8). Isolated SiRHP, when provided with suitable electron donors, can reduce SO_3^{2-} to HS^- and NO_2^- to NH_4^+ without releasing intermediates (9, 10). In contrast, isolated siroheme catalyzes these reductions inefficiently and incompletely (11). SiRHP accommodates an electron at the siroheme with a redox potential (E'_0) of -340 mV (12), and at the Fe_4S_4 cluster with an E'_0 of -405 mV (13). Reduction of SiRHP enhances substrate binding and dissociation rates 10^5 times (10), suggesting a link between cofactor electronic states and protein conformation.

The crystal structures of SiRHP presented below reveal how a protein utilizes underlying twofold symmetry to associate cofactors and enhance their reactivity for catalysis. Conservation of chemically congruent residues between symmetry-related halves of SiRHP and among other SiRs and NiRs highlights segments of sequence nec-

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