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Astronomically controlled aridity in the Sahara since at least 11 million years ago

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Abstract

The Sahara is the largest hot desert on Earth. Yet, the timing of its inception and its response to climatic forcing is debated, leading to uncertainty over the causes and consequences of regional aridity. Here we present detailed records of terrestrial inputs from Africa to North Atlantic deep-sea sediments, documenting a long and sustained history of astronomically-paced oscillations between a humid and arid Sahara from over 11 million years (Myr) ago. We show that intervals of strong dust emissions from the heart of the continent predate both the intensification of northern hemisphere glaciation and the oldest land-based evidence for a Saharan Desert by millions of years. We find no simple long-term gradational transition towards an increasingly arid climate state in northern Africa, suggesting that aridity was not the primary driver of gradual Neogene expansion of African savanna C_4 grasslands. Instead, insolation-driven wet-dry shifts in Saharan climate were common over the last 11 Myr and we identify three distinct stages in the sensitivity of this relationship. Our data provide context for evolutionary outcomes on Africa, for example, we find that astronomically-paced arid intervals predate the oldest fossil evidence of hominid bipedalism by at least 4 Myr.

Main text

Prominent from space, the Saharan desert is a vast, bare, intensely arid, dust-exporting landscape that grew 10% in response to climatic forcing during the 20th century[1]. A recognisable Saharan desert is widely suggested to originate in the late Pliocene, ~2.6 Myr ago, coupled to the development of major continental ice sheets in the Northern

Hemisphere and their expansion during the mid-Pleistocene transition (ca. 1.2–0.6 Myr ago)[2,3]. Yet, this interpretation has been challenged[4], and evidence of Saharan dust in ~4.8 Myr old Canary Island palaeosols[5] and proposed aeolian dune deposits from the Djurab of Chad dated to ~7 Myr ago[6] hint at an earlier inception of desert conditions. Weakening of the summer monsoon in climate model simulations of the response to Tethys Sea closure is hypothesized[7] to account for earlier desertification. However, the significance of the fossil dunes is highly controversial[8], estimated dates of Tethys closure span more than 20 Myr[9,10] and existing climate records are too sparse to test these competing hypotheses for desert inception. Thus, the geological history of the world's largest hot desert, which exerts a major influence on the Earth's energy balance and global biogeochemical cycles, remains poorly constrained.

Unearthing terrestrial climate history in marine sediments

We investigated the evolution of African aridity and atmospheric dust export over the last 11 Myr by developing continuous, high resolution and well-dated geochemical records of terrigenous inputs from northern Africa to marine sediment archives. We studied Ocean Drilling Project (ODP) Site 659 off Mauritania (Supplementary Figure 1), situated underneath the main modern Saharan dust plume. Pronounced cycles in colour and lithology of Site 659 sediments record changes in terrigenous content and, in the seminal study of Tiedemann et al.[11], these changes were quantified and used to infer dust inputs and aridity change on Africa over the past 5 million years. Yet, that hydroclimate reconstruction is now called into question for three reasons: First, distal, fine-grained material, likely of riverine origin, also contributes to the lithogenic fraction of sediment accumulating on the northwest African margin including Site

659[12,13] (see Supplementary Information). Second, the proportion of terrigenous material in marine sediments is also influenced by marine carbonate deposition and dissolution, which may exert a significant control, particularly in Pliocene and Miocene-aged sediments at Site 659[14,15]. Third, dust supply to the atmosphere and marine sediments is controlled not only by aridity but also wind strength[16].

We used X-ray fluorescence (XRF) core scanning to both document dust accumulation in the North Atlantic Ocean and infer African hydroclimate over the last 11 Myrs geochemically (Figure 1). Because dust flux estimates show a strong wind strength dependence and can be biased by carbonate dissolution and rapid fluctuations in sediment accumulation[15], we use two geochemical ratios, [Al+Fe]/[Si+K+Ti] and ln[Zr/Rb], to reconstruct hydroclimate variability. High [Al+Fe]/[Si+K+Ti] values are associated with low inputs of Si-rich dust and more intense chemical weathering under humid climates[17] while high ln[Zr/Rb] values indicate dominance of coarse dust over fine river clays[12]. Crucially, these two ratios employ only lithophile elements and so, unlike dust fluxes, they are independent of sea-floor carbonate dissolution and dilution and show strong agreement with hydroclimate reconstructions based upon deuterium isotopic signatures of plant waxes (Figure 2, Extended Data Figures 1–2 and Supplementary Information). Our dust flux reconstructions are similar to thorium-normalization based estimates from nearby site MD03-2705[15] (Extended Data Figure 3 and Supplementary Information) suggesting that, although the influence of dilution, dissolution, rapid sedimentation rate fluctuations and sediment remobilisation cannot be entirely ruled out, they are not likely main drivers of reconstructed dust fluxes at Site 659.

Our high-resolution records (Figure 1) reveal a long history of repeated dramatic shifts in hydroclimate between dry dusty conditions and more humid intervals reaching back 11 Myrs. The most recent of these is the African Humid Period (AHP1) in the early Holocene, when the Saharan landscape was well-vegetated and cross-cut by a network of rivers and lakes[13, 18-20] with abundant archaeological sites, some adorned with rock art depictions of giraffe, elephant, hippopotamus and domesticated cattle[21]. Throughout our record, dark terrigenous-rich sediment layers at Site 659 have significantly higher ln[Zr/Rb] and lower [Al+Fe]/[Si+K+Ti] values than pale layers indicating that they contain a greater proportion of coarser grains, and have a chemical signature approaching dust end-member values (Figure 3, Extended Data Figure 4 and Supplementary Information). Thus, the dark layers consistently signify high dust deposition linked to aridity over northern Africa rather than high riverine sediment input or merely strong sea floor carbonate dissolution. Intervening paler layers consistently record low ln[Zr/Rb] and high [Al+Fe]/[Si+K+Ti], indicating far less dusty conditions on Africa. Spectral analysis reveals that this climatic variability is primarily paced by precession and obliquity, with a strong signature of eccentricity only documented in the oldest (Miocene) part of our records (Extended Data Figures 5–6 and Supplementary Information).

Long-lived pulsing of Saharan dust export

We identify three distinct stages in the evolution of Saharan hydroclimate state over the past 11 Myrs, separated by transitions with statistically significant shifts in central tendency and dispersion in our datasets (Figure 1, Extended Data Figure 7 and Supplementary Information). Stage I (>6.9 Myr ago) reveals astronomically-paced development of arid and dust-producing regions on Africa from at least 11 Myr ago. Pronounced dark sediment layers consistently show high ln[Zr/Rb] and low [Al+Fe]/[Si+K+Ti] values, confirming that these layers are, like those in the younger part of the record, associated with aridity (Figure 3, Extended Data Figure 4). Both ln[Zr/Rb] and [Al+Fe]/[Si+K+Ti] values suggest that African climate was more arid during the Late Miocene than the Pliocene, however estimated dust flux maxima are lower. Given the importance of deflation-promoting high wind speeds (gustiness) in dust transport, we attribute this result to warmer Miocene high latitudes weakening latitudinal temperature gradients (Figure 1c), resulting in lower wind speeds and less effective deflation and dust transport[22-25]. Nevertheless, our records document substantial dust emissions that predate the oldest land-based evidence (~7 Myr ago) for Miocene initiation of northern African hyperaridity[6] by at least 4 Myr, and predate Late Pliocene intensification of Northern Hemisphere Glaciation (iNHG)[2, 3] by at least 8 Myr.

A shift towards increased [Al+Fe]/[Si+K+Ti] values followed by decreased ln[Zr/Rb] from ~6.9 Ma marks the transition into stage II (which covers the latest Miocene to the late Pliocene, ~5.75–3.5 Ma), indicating wetter conditions with more fine-grained sedimentary inputs (Figure 1f and g). Increasing variability in [Al+Fe]/[Si+K+Ti], ln[Zr/Rb] and dust fluxes is also recorded implying a heightened sensitivity of northern African humidity to astronomical forcing, although cycle amplitudes may also be more attenuated by bioturbation in Stage I where sedimentation rates are lower (Supplementary Figure 2). Dust fluxes during stage II are lower during humid events associated with insolation maxima but higher during arid insolation minima than seen for equivalent forcing in stage I. We attribute this result to the development of extensive Pliocene river systems and lakes (including Lake Chad which has a Latest Miocene origin[26]) during ‘wet orbits’ that, once desiccated, became dust-producing hot-spots[27,28] during succeeding ‘dry orbits’.

The transition to Stage III, which covers the last ~2.25 Myrs (early Pleistocene to Recent) of African climate history, is characterized by a shift towards more arid conditions (decreasing [Al+Fe]/[Si+K+Ti] centred around 3.1 Ma) followed, ca. 400 kyr later, by increased mean values of ln[Zr/Rb] and dust fluxes (which also show higher amplitude variability), closely contemporaneous with the intensification of glacials as revealed by benthic oxygen isotope records[29] (Figure 1b). The highest dust fluxes in our records during Stage III are consistent with the suggestion that the growth of large continental ice sheets in the Northern Hemisphere promoted the development of more arid and dusty conditions on Africa through steepening latitudinal temperature gradients and strengthening winds[3,30].

Consistency of source

We investigated the source of terrigenous material accumulating at ODP Site 659 over the last 11 Myr using radiogenic isotopes. Today, dust is generated widely across three geochemically distinct northern African preferential source areas (PSAs)[31]. Strontium and neodymium isotopic signatures of the lithogenic fraction of Site 659 sediments are remarkably consistent compared to the range in modern PSA signatures and lie between those reported for the Central and currently undersampled Western source regions[31] (Figure 4). These results imply that the terrigenous source regions remained broadly unchanged over the past 11 Myr and that variability in grain size is not a major control on our data (Extended Data Figures 8–10 and Supplementary Information). This observation, together with the high ln[Zr/Rb] and [Al+Fe]/[Si+K+Ti] values recorded during Miocene insolation minima, indicates that dust-producing regions existed in the inland Sahara region and were activated by astronomical forcing back to at least 11 Myr ago. We conclude that while iNHG in the late Pliocene (~3.3–2.5 Myr ago) drove increased dust fluxes (Figure 1h), it was not responsible for the inception of the Saharan desert.

Rainfall climate, C_4 savannas and evolutionary outcomes

Savannas dominated by C_4 grasses expanded extensively during the late Miocene[32,33], but the cause of this major reorganization of the terrestrial biosphere is controversial. Both aridification and decreasing $p\mathrm{CO}_2$ are proposed drivers of this grassland ecosystem shift, aided by strong positive feedbacks associated with fire, and regionally modified by latitude, altitude and rainfall seasonality[32-37]. To better understand the potential role of hydroclimate variability in driving the expansion of C_4 grasslands, we compare our geochemical datasets to stable carbon ($\delta^{13}C$) and hydrogen (δD) isotope signatures of leaf wax long-chain *n*-alkanes at Site 659 (Figures 1d and 2). The carbon isotope signature of plant waxes is strongly controlled by the photosynthetic pathway of the parent plant: *n*-alkanes from C_4 plants are isotopically heavier (–21.4±2.2 ‰) than C_3 plants (–34.9±2.7 ‰)[38].

We find little similarity between the gradational expansion of C_4 grasses in northern Africa and long-term hydroclimate trends (Figure 1). The plant wax records integrate material from across a large source area that was dominated by C_4 vegetation during the Quaternary[39,40], potentially contributing to the expression of only modest change in *n*-alkane $\delta^{13}C$ in response to insolation forcing in the youngest part of the record. Nevertheless, the hydroclimate signal in North Africa ([Al+Fe]/Si+K+Ti], ln[Zr/Rb] and *n*-alkane δD[36,39,40]) is dominated by astronomical variability, while the vegetation signal (*n*-alkane $\delta^{13}C$[36,39,40]) is dominated by the gradual long-term trend that defines the C_3-C_4 transition and is not replicated by the hydroclimate reconstructions (Figures 1 and 2, Supplementary Figure 3). Therefore, we find that a simple hydroclimate driver cannot explain C_4 grassland expansion in northwest Africa through the Neogene. A similar conclusion was reached by ref. 36. However, while that study ruled out a hydroclimate control because of little inferred change in regional rainfall, we document major changes in hydroclimate unaccompanied by a strong response in C_4 plant dominance (Figure 2). This decoupling between aridity and C_3-C_4 composition does not rule out an influence of aridity on C_4 vegetation dominance, but suggests that the relationship was not simple. An improved record of Neogene pCO_2 variability (Figure 1a) is needed to better understand coupling between global climate state, hydroclimate and the rise of C_4 grasslands.

An inferred progressive aridification of Africa and spread of savanna ecosystems has been linked to speciation and development of long-range mobility in hominins[3, 41] and extinction of many large mammals[32,42]. Our long, detailed, continuous marine records help to develop the environmental framework needed to assess these evolutionary outcomes on land, including the hypothesized relationships between habitat instability and development in hominins of characteristics that promote adaptability[43,44]. We highlight the longevity and primacy of the astronomical signal in controlling hydroclimate in northern Africa. Our findings of three distinct phases in North

African hydroclimate, including relatively dry conditions during the warm Miocene, suggest no simple escalation in aridity through the late Cenozoic associated with global cooling, and argue against aridity as the primary driver of C_4 grassland expansion. Our records show evidence of changes in mean state and/or amplitude of variability in hydroclimate close to the oldest suggested fossil evidence of hominin bipedalism[45] and the earliest records of the genus *Homo*[46] (Figure 1). However, the confidence interval on the origination of the principal taxa, especially suggested bipeds, is large, implying earlier emergence[47]. New fossil finds will inevitably revise the hominin chronology, enabling a stronger understanding of the evolutionary link to climate. Meanwhile, our study reveals astronomically-paced dry and dusty conditions on Africa long before both the oldest land-based evidence of a Saharan desert and the intensification of Northern Hemisphere glaciation.

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Author contributions

A.J.C., P.A.W., C.P.O. and D.J.B. designed the study. A.J.C. and T.W. performed the XRF measurements and developed the age model. B.D.A.N. generated the *n*-alkane $\delta^{13}C$ data and M.J.C. and A.J.C. generated the radiogenic isotope data. A.J.C. performed the sediment endmember unmixing. A.J.C. and P.A.W. led the analysis and interpretation of results with input from all authors. A.J.C., P.A.W., D.J.B. and C.P.O. led the writing of the manuscript with contributions from all other authors.

Competing interests

The authors declare no competing interests.

Figures

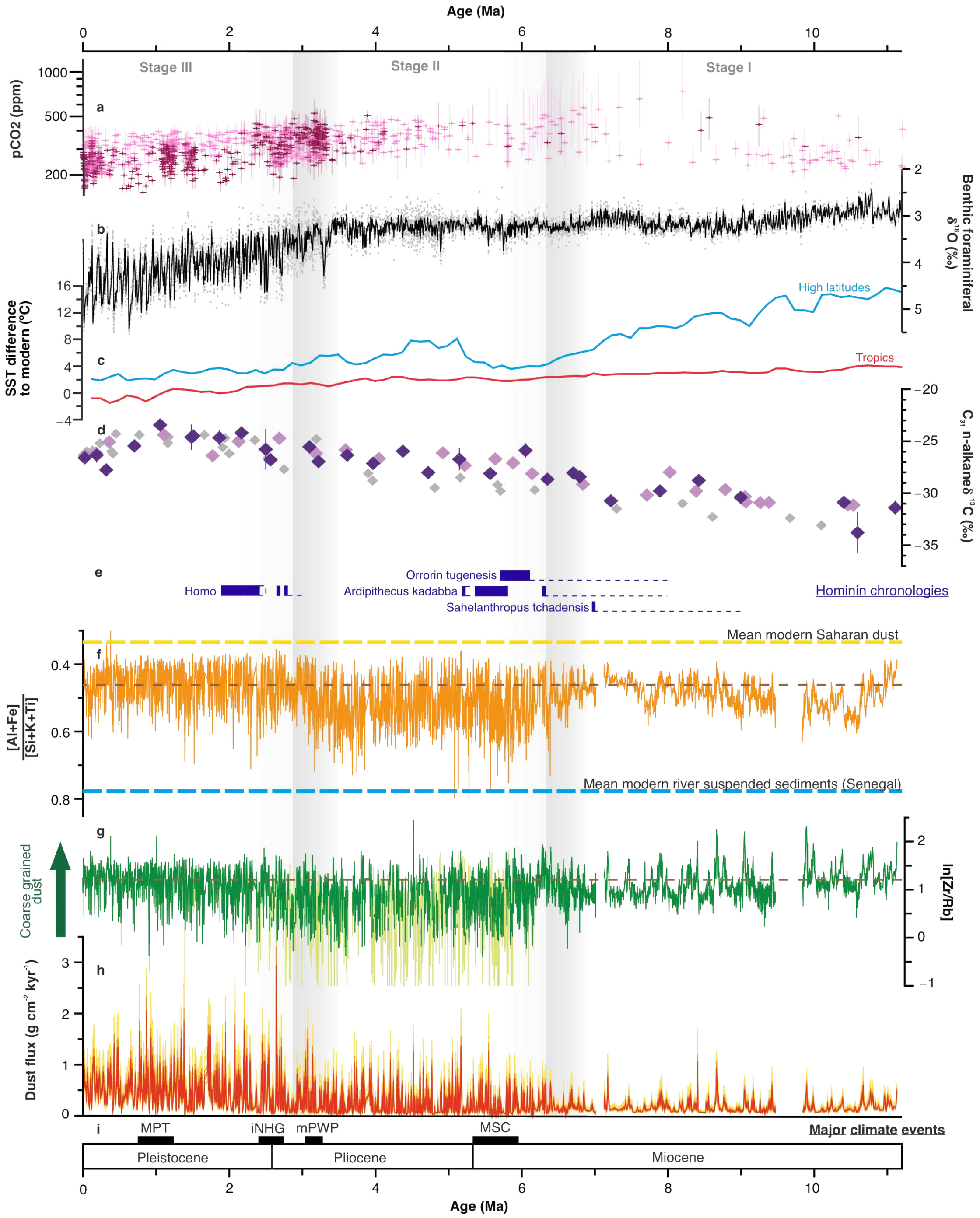

Figure 1| African hydroclimate compared to globl change over the past 11 Myr. a Atmospheric pCO_2 reconstructions (references in Supplementary Information), purple: estimates from planktonic foraminiferal boron isotopic signatures, pink: alkenone-derived estimates. **b** Cenozoic global reference benthic foraminfera oxygen isotope dataset, with 20 kyr smoothing[29]. **c** Regional stacks of sea surface temperature difference to modern annual means[23], blue: >50°N, red: tropics. **d** $\delta^{13}C$ signature of *n*-alkanes from Site 659 (dark purple: this study, light purple: ref. 36) and offshore E. Africa[48] (grey). Error bars indicate 1σ. **e** Major events in hominid evolution. Solid bars: taxon ranges, dashed lines: confidence interval on taxon origin[47]. **f** [Al+Fe]/[Si+K+Ti] of calibrated elemental abundances. Modern endmember values updated from ref. 17 (see Supplementary Information). Dashed line marks mean over last 1 Myr. **g** ln[Zr/Rb] XRF core scan ratios. Dashed line marks mean over last 1 Myr. **h** Site 659 estimated dust flux (3pt smoothed). Median value in red; 1%, 5%, 25%, 75%, 95% and 99% percentiles also shown in shades of orange/yellow. **i** Major global climate events. MPT: mid-Pleistocene transition, iNHG: intensification of Northern Hemisphere glaciation, mPWP: mid-Pliocene warm period, MSC: Messinian Salinity Crisis. Vertical grey shading indicates transitions between climate stages.

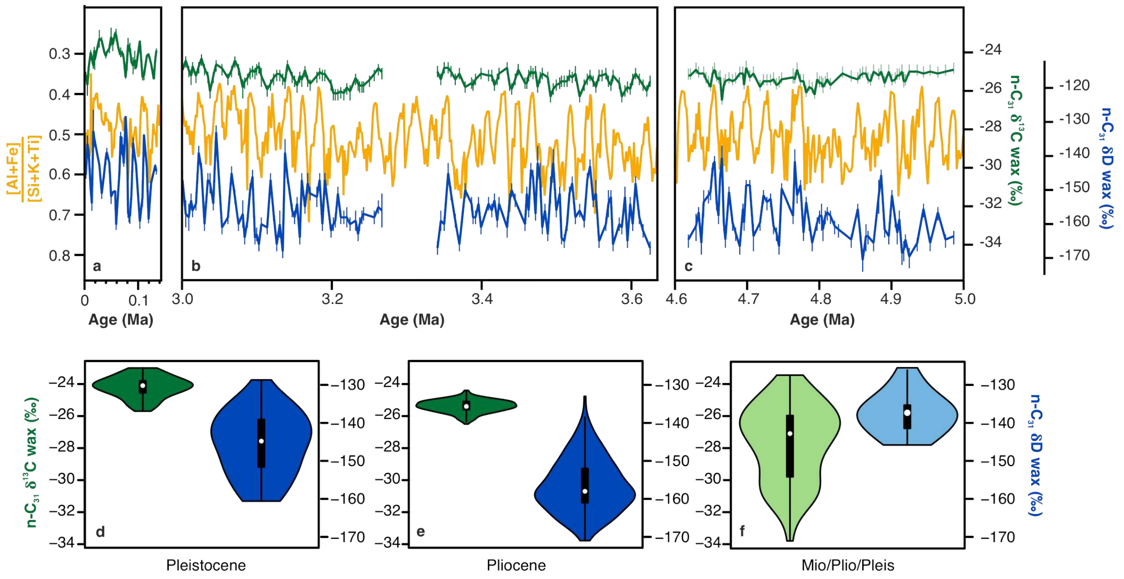

Figure 2| Relationship between lithophile element ratios and hydrogen and carbon isotopic compositions of plant waxes at Site 659. a Last glacial cycle, **b** late Pliocene, **c** early Pliocene. [Al+Fe]/[Si+K+Ti] of bulk sediment in orange (this study). Leaf wax C_{31} *n*-alkane $\delta^{13}C$ data in green[39,40] and δD in blue[39,40], error bars mark 1 standard deviation. High [Al+Fe]/[Si+K+Ti] and low *n*-alkane δD values both indicate more humid conditions. **d-f** Violin plots indicating the distribution of leaf wax C_{31} *n*-alkane $\delta^{13}C$ (green) and δD (blue**)** data in high-resolution snapshots in the Late Quaternary (**d**) and Pliocene (**e**) (data from refs. 39 and 40), and low-resolution records covering the last 11 Myr (**f,** data from this study and ref. 36). White dots indicate median values, black rectangles indicate interquartile range.

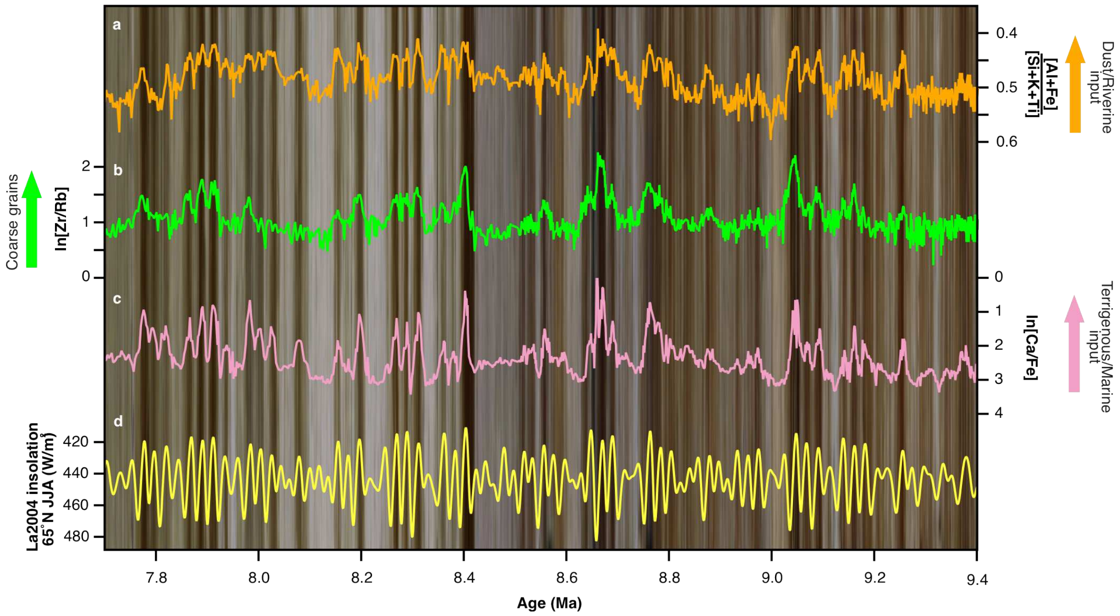

Figure 3| Strong response of African hydroclimate to astronomical forcing recorded at Site 659. Example from the late Miocene. **a** [Al+Fe]/[Si+K+Ti] of calibrated elemental abundances. **b,c** ln[Zr/Rb] and ln[Ca/Fe] XRF core scan ratios. **d** Summer insolation at 65°N[49]. Background colour shows the composited sediment core image. Darker sediments signal dry/dusty conditions, lighter sediments signal more humid intervals (see also Extended Data Figure 4 and Supplementary Information).

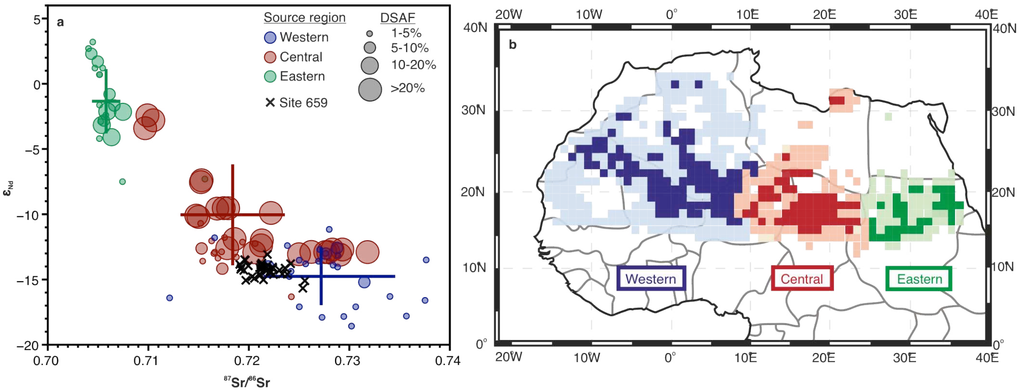

Figure 4| Radiogenic isotope signature of Site 659 lithic fraction compared to values of preferential source regions (PSAs) reveals consistent source. a Site 659 bulk sediment data (black crosses) compared to source region measurements (circles) coloured by PSA[31]. Data point size indicates dust source activation frequency (DSAF)[28]. Crosses indicate mean signatures of each PSA weighted by annual DSAF, bars denote ± one weighted standard deviation[31]. Samples for which only ε_{Nd} or $^{87}Sr/^{86}Sr$ data exist are included in the calculation of mean values but are not plotted. **b** Map of North African PSAs, adapted from ref. 31. Dust sources with activation frequencies >5% shown in bold colours, <5% in pale colours.

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Methods

Site Location

ODP Site 659 is situated in the tropical North Atlantic Ocean on the Cape Verde Rise, offshore Mauritania (18.077°N 21.026°W, 3070 m water depth, for map see Supplementary Figure 1). Today, this region receives dust delivered by the trade winds during winter and spring, with additional dust transported over the site by the Saharan Air Layer predominantly during boreal summer[50-56]. The Algeria-Morocco region, Mali, Mauritania, Libya, Niger, Western Sahara and the Sahel have all been identified as regions that may currently contribute dust to the Cape Verde Islands and the surrounding ocean sediments, while evidence for a major contribution from the Bodélé depression is weaker[50-53,56-65]. The elevated position of Site 659 on the Cape Verde Rise protects it from major mass transport deposits, although grain size data reveal that multiple sites along the northwestern African margin including GeoB7920[12] and Site 659 received a significant fine-grained lithogenic component during both the Holocene and last glacial cycle that is distinct from modern dust inputs and is often attributed to a distal influence from African (palaeo)rivers[12] (Extended Data Figures 9–10 and Supplementary Information).

X-ray fluorescence

Data were collected from a total of about 275 m of drill core from three holes at ODP Site 659 using the XRF Core Scanner II (AVAATECH Serial No. 2) at MARUM - University of Bremen. Measurements were taken every 1–5 cm down core to give a consistent approximate temporal resolution of 1.5–2 kyr. Count times were 20 seconds and generator settings of 30 and 10 kV with currents of 0.75 and 0.2 mA respectively were used. The split core surface was covered with a 4 micron thin

SPEXCerti Prep Ultralene foil to avoid contamination of the XRF measurement unit and desiccation of the sediment. The data were acquired using a Canberra X-PIPS Silicon Drift Detector (SDD; Model SXD 15C-150-500) with 150eV X-ray resolution, the Canberra Digital Spectrum Analyzer DAS 1000, and an Oxford Instruments 50W XTF5011 X-Ray tube with rhodium (Rh) target material. Raw X-ray spectra were processed using an iterative least square software (WIN AXIL) package from Canberra Eurisys. Repeat runs of several core sections were used to correct for any drift in counts over time.

Ca/Fe ratios provide an indicator of the relative proportions of marine (dominated by biogenic calcium carbonate) and terrigenous (the main source of iron) sediment (Supplementary Figure 4). We use two geochemical ratios to determine the relative contribution of aeolian material to the sediment. Modern Saharan dust has much lower [Al+Fe]/[Si+K+Ti] values than samples of suspended sediment from the Senegal River (Figure 1f), the nearest and largest active river to Site 659, which drains deeply weathered lateritic tropical soils much richer in Al and Fe than the Si-rich Sahelian/Saharan dust sources[17]. We also employ Zr/Rb as an independent grain size proxy because the lithogenic dust fraction is significantly coarser than riverine-derived material on the northwest African margin[12,66], and contains a higher proportion of Zr-bearing zircon grains than Rb-rich river clays[67]. Additional geochemical ratios are shown in Supplementary Figures 5–6. Our XRF records have an average temporal resolution of approximately 2000 years.

A suite of discrete sediment samples from ODP Site 659 incorporating a wide range of lithologies were analysed by energy dispersive polarised x-ray fluorescence spectrometry (EDP-XRF) at the University of St Andrews to convert the semi-quantitative XRF scanner counts into concentrations (Supplementary Figure 7). The downcore records plotted are calculated from ratios of these calibrated values, except for ln[Zr/Rb] where counts of these trace elements are plotted.

Stratigraphy

We created a new spliced composite section for ODP Site 659 to a depth of 203.17 revised metres composite depth (rmcd) by correlating both the XRF core scan data and core images between the three holes (A–C) drilled at Site 659, using the Code for Ocean Drilling Data macros[68]. An age framework was developed from paleomagnetic, nannofossil and planktonic foraminiferal datums[14,69] with ages updated to incorporate more recent improvements in astrochronology[70-72]. We then correlated previously published oxygen isotope values of the benthic foraminifera *Cibicides wuellerstorfi* at Site 659[11] transferred onto our composite depth scale to the LR04 benthic oxygen isotope stack[73] to improve age control for the youngest 5 Myr. The final age model was then generating by tuning our XRF count data (ln[Ca/Fe]) to summer insolation calculated at 65°N in the La2004 astronomical solution[49], guided by the framework provided by the biostratigraphic, magnetostratigraphic and benthic oxygen isotope record. The construction of this age model is illustrated in Supplementary Figure 8. In Supplementary Figure 2, we compare the age-depth relationship of our astronomically-tuned age model to existing bio- and magnetostratigraphic datums. Continuous wavelet power spectra[74] of both astronomically-tuned and non-astronomically tuned time series data are shown in Extended Data Figure 5, with results from REDFIT[75] spectral analysis shown in Extended Data Figure 6.

Geochemical end-member unmixing

Dust fluxes were estimated using an end-member unmixing approach to deconvolve the relative proportions of different contributions to the bulk marine sediment, largely following the method of Mulitza et al. (2010)[17]. In this approach, bulk sediment is assumed to consist of three components: aeolian, riverine and marine inputs. The relative abundances of Al, Si, Fe, K, Ti and Ca concentrations at Site 659 (calibrated by a multivariate log calibration method[76,77] using the AvaaXelerate software[78]) were compared to the chemistry of endmember compositions estimated from analyses of modern sediments. The riverine endmember was constructed from ten measurements of suspended sediment from the Senegal River[79], the present day catchment of which stretches both north and south of Site 659. A total of 48 measurements were used to construct a dust endmember, covering a wide geographical area and incorporating data from dust traps, atmospheric sampling and bulk sediment from known dust source areas with a range of grain sizes and local climates[59,62,80-91]. A marine endmember composition of 2% Si and 98% Ca was assumed[17]. To incorporate the effect of temporal changes in active terrigenous source regions and lithic fraction chemistry into our dust flux estimates, a bootstrapping approach was applied. During every realization, 10 of the 48 dust compositions and 4 of the 10 river compositions were selected by a bootstrap with replacement routine, with the unmixing procedure carried out for a total of 500 realizations of endmember compositions. Proportions of aeolian and riverine endmembers were converted to fluxes using sedimentation rates calculated from our composite depth scale and age model, and dry bulk sediment densities estimated from shipboard measurements of gamma-ray attenuation[14] which were calibrated using the discrete dry bulk density measurements of Tiedemann (1991)[92]. We show that there is good agreement between our dust flux estimates and those based on ^{230}Th normalization[15] and lithogenic %[11] in Extended Data Figures 2–3, Supplementary Figure 4 and Supplementary Information.

Lithogenic grain size measurements and geochemical data strongly support the concept of distal riverine inputs to Site 659. The proportion of the terrigenous fraction that is attributed to aeolian inputs by this geochemical unmixing approach varies strongly down core, with estimates of 0–87% dust origin. These estimates are broadly in line with those of Tjallingii et al. (2008)[12] who find that dust accounts for 5–95% of the lithic fraction through the last glacial cycle at nearby site GeoB7920 using a unmixing approach based upon grain size distributions. We cannot, however, rule out contributions to the fine-grained, high Al+Fe end-member from very fine dust or resuspended shelf material. Thus, our dust fluxes are minimum (conservative) estimates (see Supplementary Information).

Radiogenic isotopes

The Sr and Nd isotope composition of terrigenous sediments from ODP Site 659 were measured at the University of Southampton National Oceanography Centre. Approximately 1g of crushed and homogenised dried bulk sediment was decarbonated using 10 % acetic acid solution for 24 hours on a shaker table and then rinsed in deionized water. Samples were placed in a 5 % hydrogen peroxide solution for 48 hours at 65 °C to remove organic matter, then rinsed three times in deionized water. 1M $MgCl_2$ was added to the samples to remove any Sr adsorbed from seawater, which were shaken overnight then rinsed three times. A buffered hydroxylamine HCl, Na-EDTA and acetic acid solution was added to remove authigenic coatings, with samples mixed for 24 hours and then rinsed. A magnesium ion solution was added to aid flocculation if a colloid phase formed at any stage during processing. The ‘cleaned’ sediments were then totally digested in concentrated nitric acid (HNO_3) and hydrofluoric acid (HF), followed by 6M HCl.

Digested sediment solutions for Sr isotope analysis were purified on columns filled with Sr-spec Resin in 3M HNO_3. Samples were loaded onto tantalum (Ta) filaments with a 1M HCl solution after a tantalum chloride (TaCl) activator solution was loaded. Isotopic measurements were made by thermal ionization mass spectrometry (Thermo-Fisher TRITON Plus). Repeated measurements of the Sr isotope standard NBS987 gave $^{87}Sr/^{86}Sr = 0.710243 \pm 0.000021$ (2σ).

Neodymium was purified from the digested sediments for isotope analysis using a standard column chemistry procedure, based upon the methods of Cohen et al. (1988)[93]. Cation exchange resin was used to strip iron and titanium from the samples, then the remaining material was then run through using Ln-Spec^{TM} resin columns[94] to purify and concentrate neodymium. Neodymium isotope ratios were measured by multi-collector inductively coupled plasma mass spectrometry (Thermo NEPTUNE). Instrumental mass bias was corrected using the procedure of Vance and Thirlwall (2002)[95], adjusting to a $^{146}Nd/^{144}Nd$ ratio of 0.7219 and using cerium-doped standards to correct for interference of ^{142}Ce on ^{142}Nd. All ratios were normalised to the JNdi-1 standard ($^{143}Nd/^{144}Nd = 0.512115 \pm 0.000007$)[96], with independent measurements of JNdi-1 = 0.512115 ± 0.000006 (2σ). Isotopic signatures are expressed in epsilon notation, relative to the chondritic uniform reservoir value of 0.512638[97].

Leaf wax $\delta^{13}C$

Compound specific stable carbon isotopes ($\delta^{13}C$) of the C_{31} and C_{33} *n*-alkane were obtained from 30 samples from ODP Site 659 at the Organic Geochemistry Unit, University of Bristol. For this purpose, between 5 and 10 gram of freeze-dried sediment was extracted using a Microwave extraction system (Milestone Ethos Ex) and a mixture of dichloromethane (DCM) and methanol (MeOH) (9:1, v/v). Fine grained sediment was removed by eluting the total lipid extract (TLE) over a short (4 cm) inert $NaSO_4$ column using 4 ml of DCM:MeOH (9:1). The TLE was dried under a gentle flow of N_2. The TLE was subsequently derivatized using pyridine and BSFTA (1 hr at 70 °C) and within 24 hrs analysed on an Isoprime 100 gas chromatograph combustion isotope ratio mass spectrometer (GC-C-IRMS). Samples were measured in triplicate and $\delta^{13}C$ values converted to VPDB by bracketing with an in-house gas (CO_2) of known $\delta^{13}C$ value. Instrument stability was monitored by regular analysis of an in-house fatty acid methyl ester standard mixture and indicates that long-term instrument stability was better than ± 0.3 ‰. Injection volume was 1 or 2 μl onto to a Zebron-I nonpolar column (50 m x 0.32 mm i.d., 0.10 μm film thickness). The GC oven was programmed as: 70 °C (1 min hold) to 130 °C at 20 °C min^{-1}, then to 300 °C at 4 °C min^{-1}, and a final hold at 300 °C for 25 min. Samples were automatically integrated using the Ion Vantage software.

For some samples, the concentration of the C_{33} *n*-alkane was not enough to obtain a reliable $\delta^{13}C$ value. The $\delta^{13}C$ of the C_{31} *n*-alkane was consistently lighter than that of the C_{33} *n*-alkane from the same sample by on average 2 ‰.

Lithogenic grain size distributions at ODP Site 659

The distribution of grain sizes in lithogenic material can be used to identify different terrigenous components in marine sediment. 55 samples from ODP Site 659 were selected to generate grain size distribution data, incorporating a range of sediment ages and lithologies. Calcium carbonate, authigenic coatings and organic matter were removed following the procedure used for radiogenic isotope analysis. In addition, a biogenic silica removal step was applied by sonicating samples in 1.5M NaOH. Calgon was then added to the samples to prevent flocculation before analysis of the lithogenic grain sizes using a Laser Coulter Sizer at the University of Southampton Waterfront Campus, NOCS. The results of these grain size analyses are shown in Extended Data Figure 9d.

The signatures of different terrigenous sources were separated from the bulk sediment signature following the end-member modeling analysis approach (EMMA) of Dietze et al. (2011)[98]. A two lithogenic end-member solution explains 88% of the variance in the Site 659 data set, with the end-members shown in Extended Figure 9. The coarser of these two endmembers shows a strong match with measurements of African dust (Extended Data Figure 9a-b)

Data availability

The data presented in this study are available in the Zenodo repository (DOI: 10.5281/zenodo.6594643).

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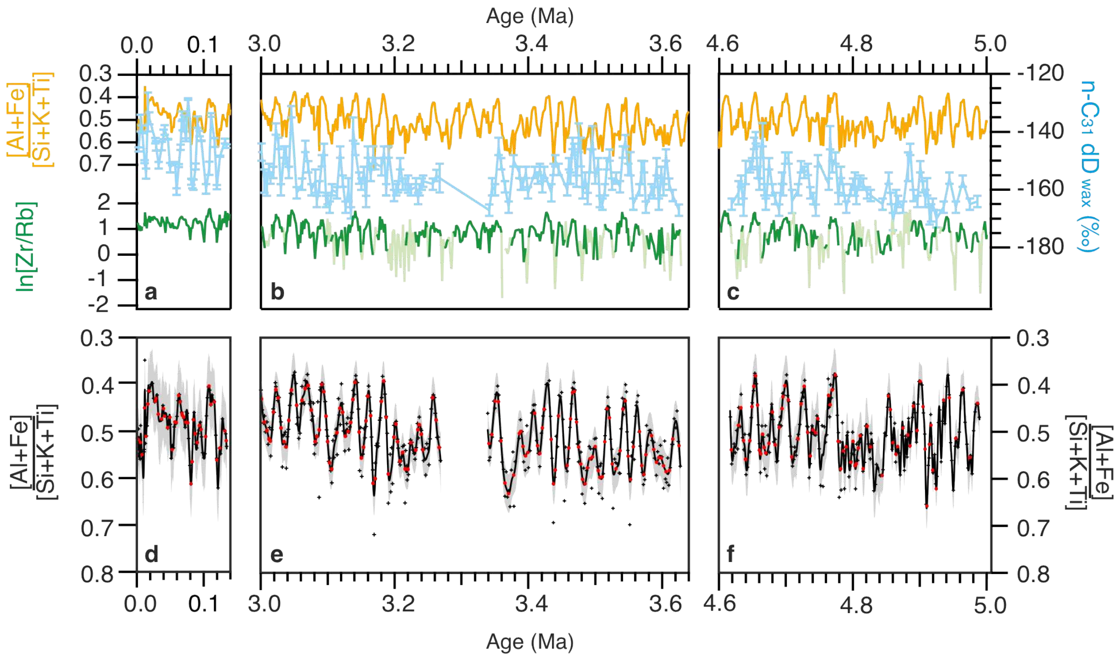
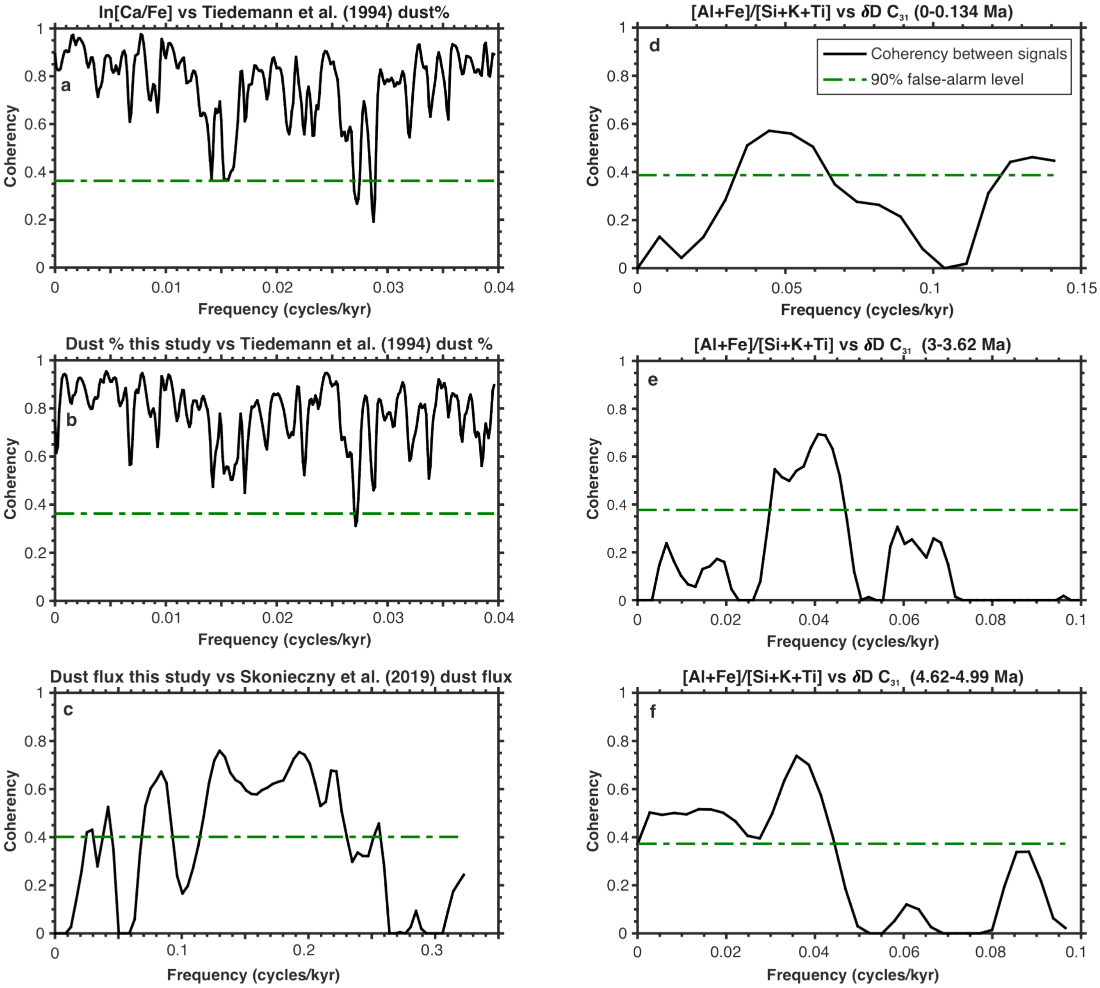
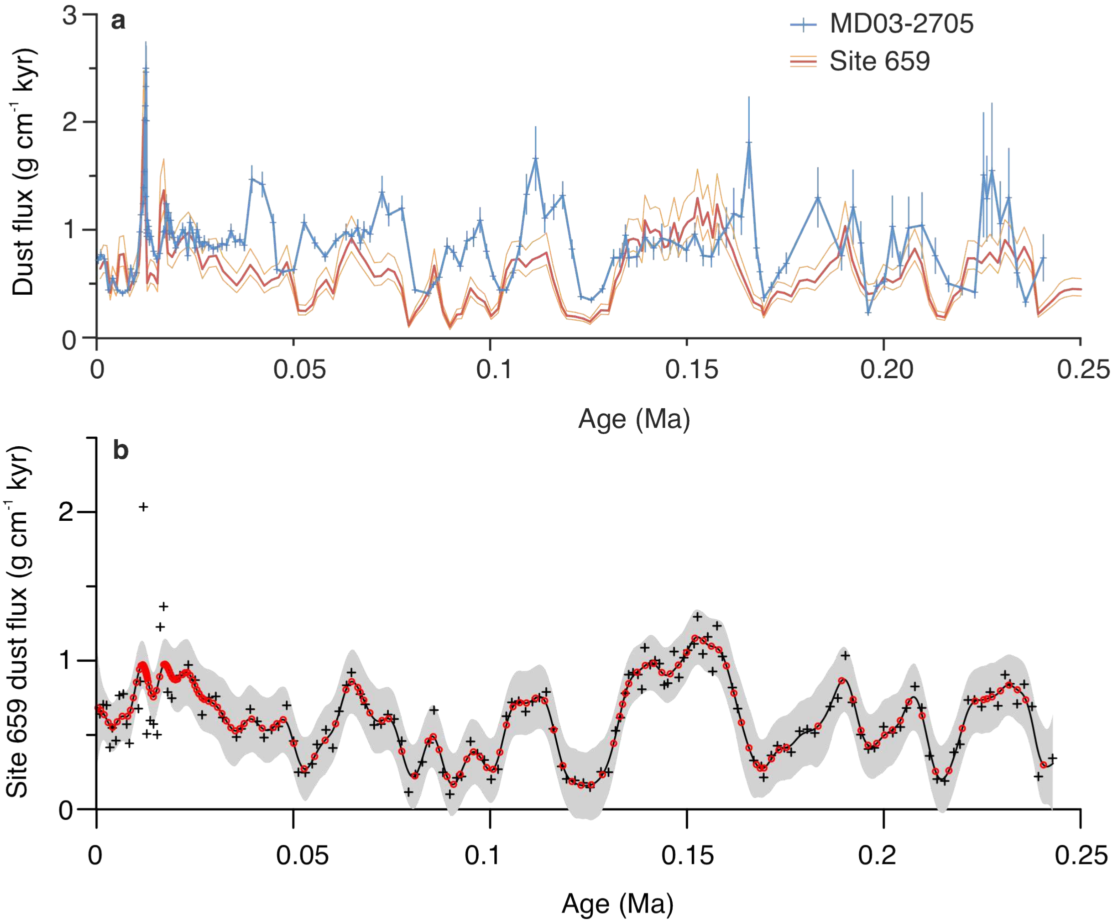


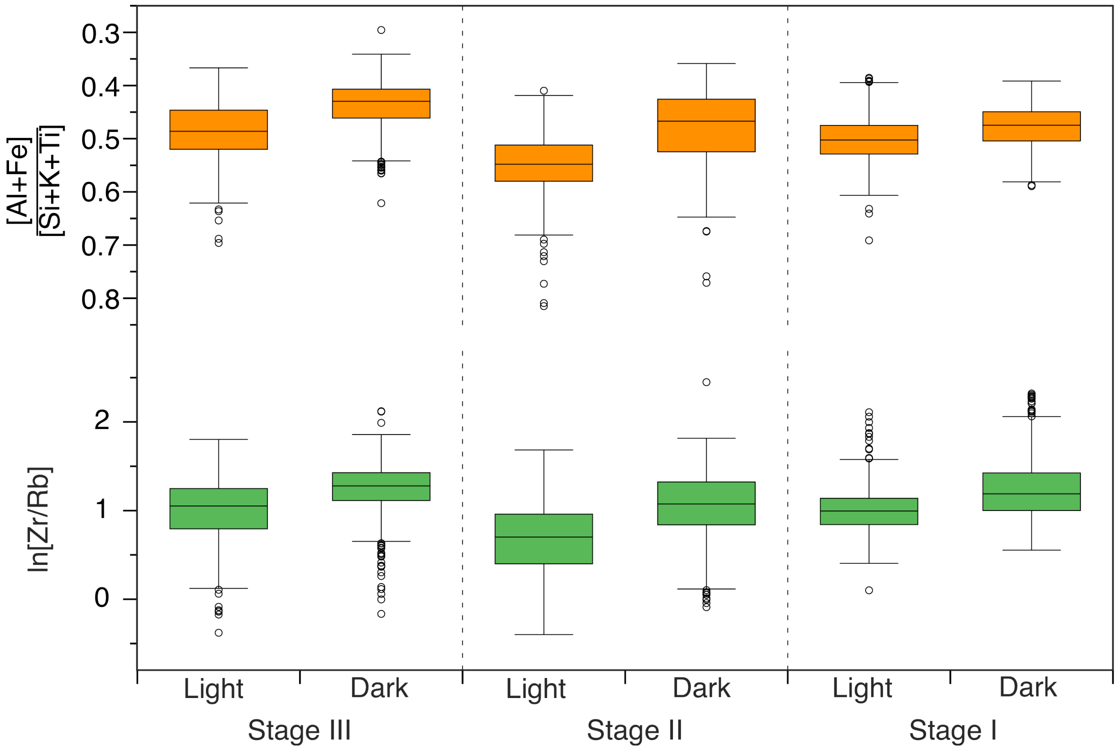
Extended Data Figure 1| Comparison between hydroclimate proxies measured at Site 659. a, b, c Bulk sediment [Al+Fe]/[Si+K+Ti] (orange), δD signatures of C_{31} *n*-alkanes (pale blue, 1σ error bars)[39,40], ln[Zr/Rb] (green, pale colours indicate low element counts). **d, e, f** Generalised additive model (GAM) fit of Site 659 [Al+Fe]/[Si+K+Ti] data shown by black line with grey shaded confidence band (2 standard error). Black crosses indicate original data points with resampled data points used in Kendall's tau-b correlation tests indicated by red circles (see Supplementary Information). **a,d** late Pleistocene, **b,e** late Pliocene, **c,f** early Pliocene.



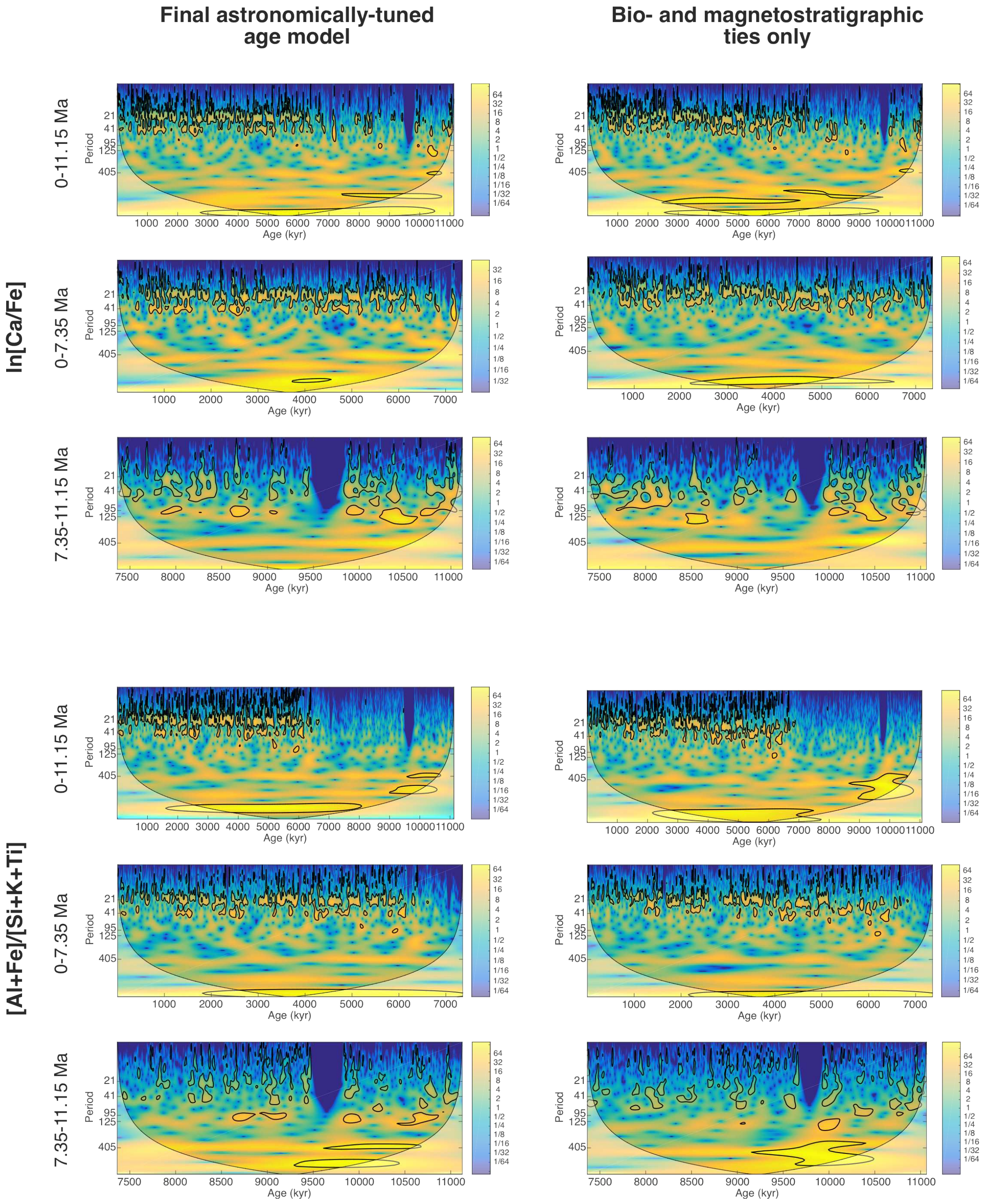
Extended Data Figure 2| Coherency spectra comparing our data from Site 659 to published dust and hydroclimate records from the same and nearby sites. Green dashed line marks 90% Monte Carlo false-alarm level. **a** & **b** Coherency between the dust % estimates from Site 659 of Tiedemann et al. (1994)[2] and our ln[Ca/Fe] (**a**) and dust flux (**b**) estimates over the last 8 Myr. **c** Coherency between our estimated dust fluxes and those of Skonieczny et al. (2019)[15] from nearby site MD03-2705 over the last 240 kyr. **d**, **e**, **f** Coherency between our [Al+Fe]/[Si+K+Ti] values and C_{31} *n*-alkane δD values from Site 659 from Kuechler et al. (2013, 2018)[39,40] for three time slices in the Quaternary (**d**) and Pliocene (**e**, **f**).



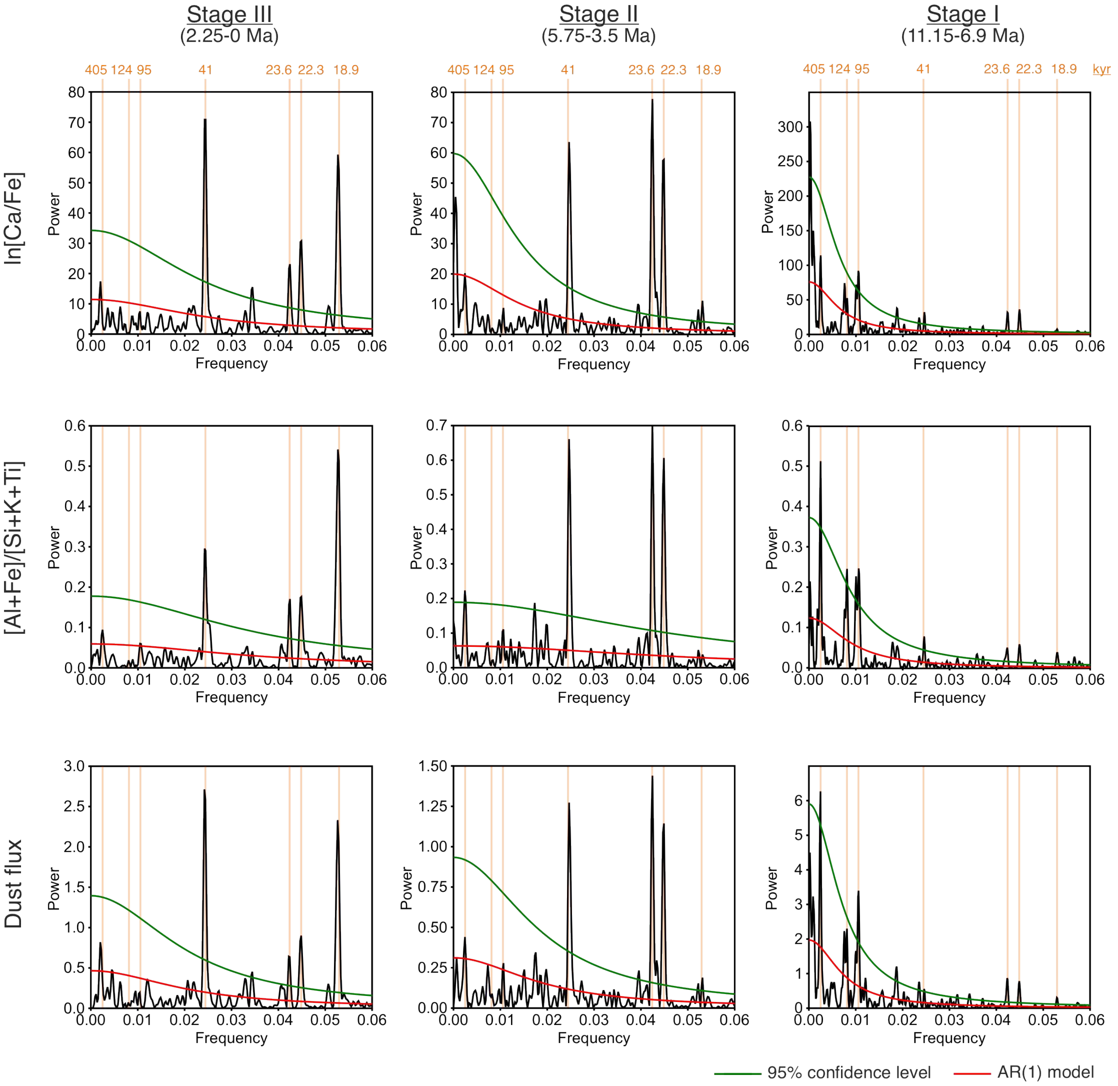
Extended Data Figure 3| Comparison between methods for calculating dust fluxes to marine sediments over the last 250 kyr. a Red: modal dust flux estimates from Site 659 based on a geochemical end-member unmixing approach, with orange lines marking ± 1 standard deviation of 500 realizations. Blue: Dust flux estimates from site MD03-2705 (directly adjacent to Site 659) calculated by ^{230}Th normalization[13] with error bars indicating ± 1 standard deviation. **b** Generalized Additive Model[99] fit of Site 659 median dust fluxes shown by black line with grey shaded confidence band (2 standard error). Black crosses indicate original data points with resampled data points used in Kendall's tau-b correlation tests indicated by red circles (see Supplementary Information).



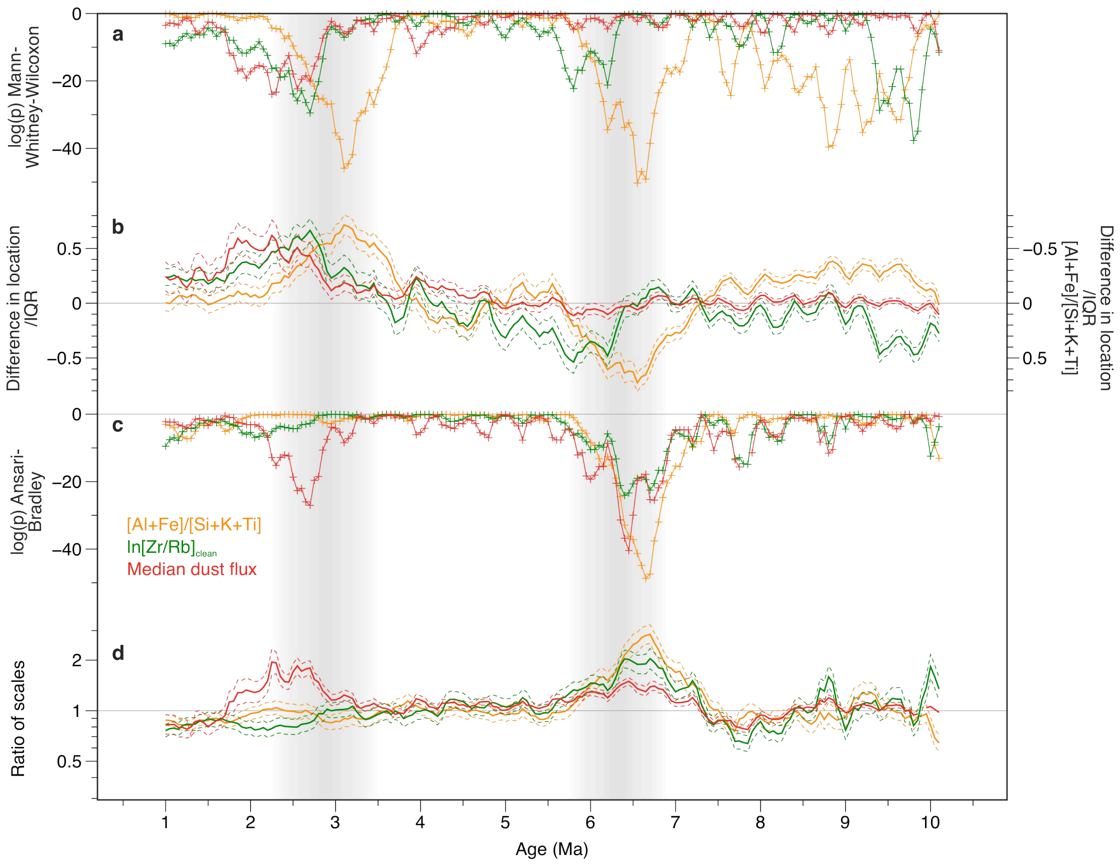
Extended Data Figure 4| Box and whisker plot illustrating co-variation in sediment colour and geochemistry at Site 659. Top: [Al+Fe]/[Si+K+Ti], bottom: ln[Zr/Rb]. Data are plotted from "light" and "dark" sediment layers (as defined by the method described in the Supplementary Information) for each of the three time stages (Stage I: 11.15 – 6.7 Ma, Stage II: 5.75 – 3.5 Ma, Stage III: 2.25 – 0 Ma). Box indicates interquartile range (IQR) with line marking median value, outliers (>1.5 IQR from median) marked with circles and whiskers drawn to the maximum/minimum values excluding outliers. Mann-Whitney-Wilcoxon tests were used to test the null hypothesis that samples from light and dark layers have identical continuous distributions with equal medians for each time interval. All resulting *p*-values were <0.001, giving >99.9% confidence that the differences between light and dark layers are significant.



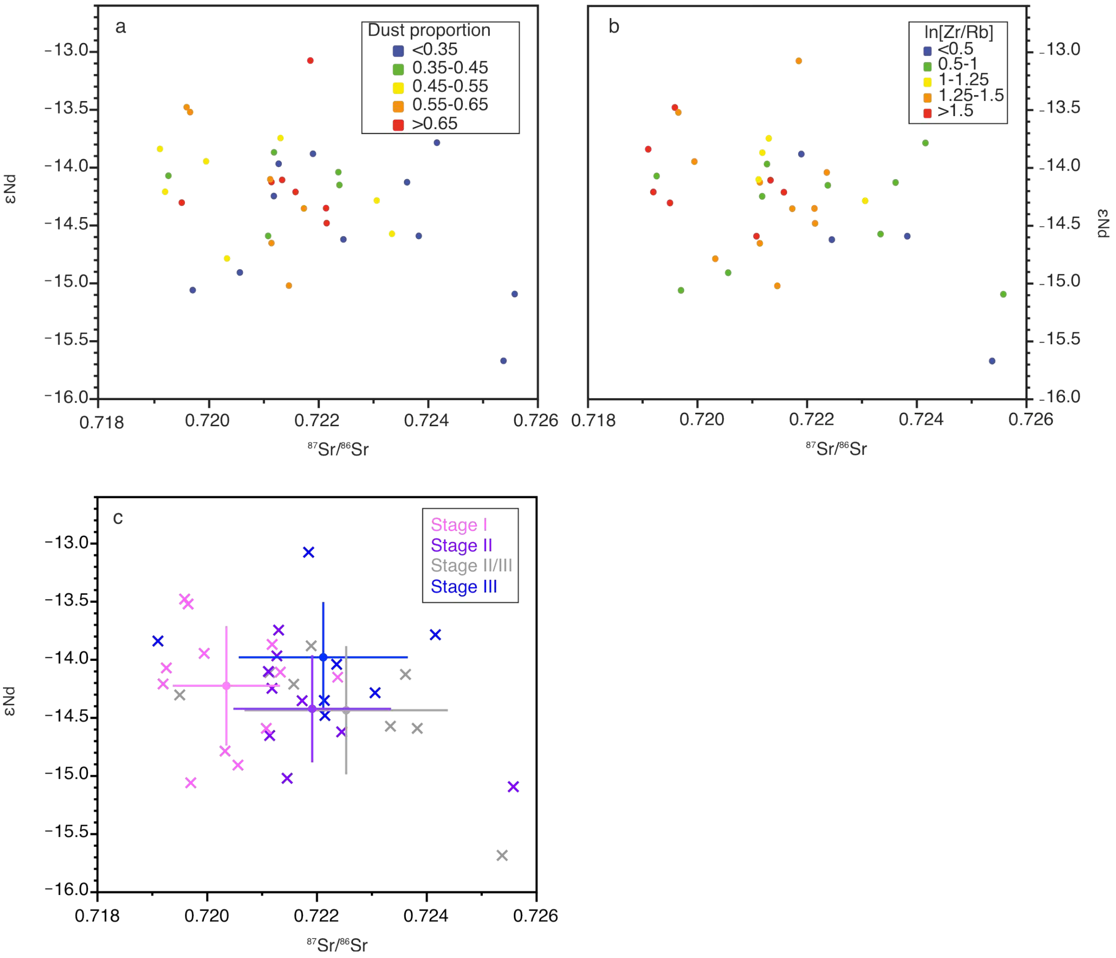
Extended Data Figure 5| Wavelet analysis of Site 659 geochemical records. Comparison between continuous wavelet power spectra of calibrated ln[Ca/Fe] (top) and [Al+Fe]/[Si+K+Ti] (bottom) data on the astronomically-tuned age model (left) and an untuned age model based solely on biostratigraphic and magnetostratigraphic datums[14,69] (right). Thick black contours designate the 5% significance level against red noise and the cone of influence is shown as a lighter shade, where edge effects may cause distortion. Data were detrended and smoothed (5-pt moving average) prior to the wavelet analyses. Separate spectra were also generated for the older and younger sections of the full record to reduce the impact of temporal changes in cycle amplitude on the detected frequencies. Analyses were performed and figures generated using the Matlab code of Grinsted et al. (2004)[100]. See Supplementary Information for further discussion.



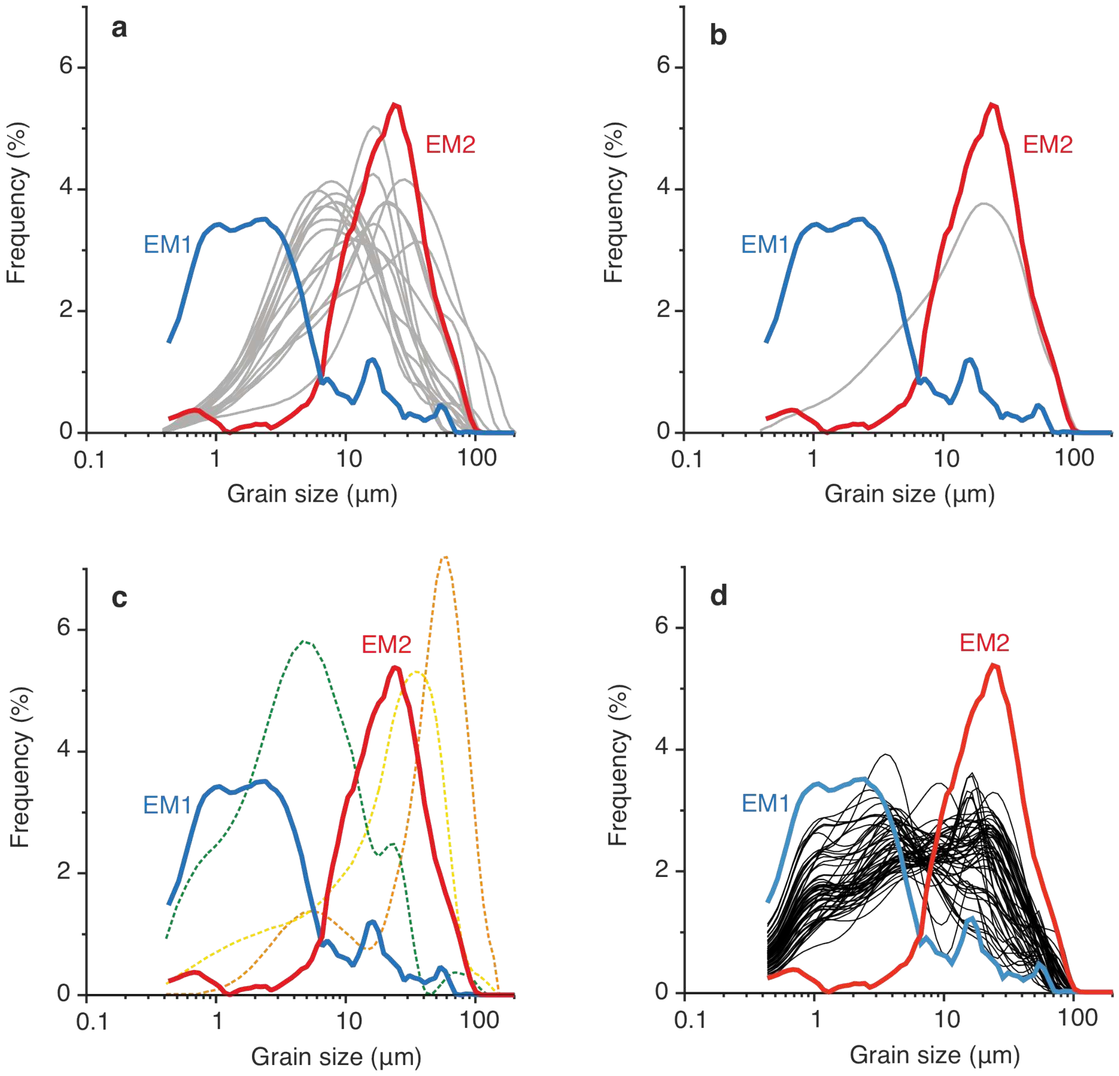
Extended Data Figure 6| REDFIT spectral analysis[75] of Site 659 geochemical records. Top: ln[Ca/Fe] ratios, middle: calibrated [Al+Fe]/[Si+K+Ti] ratios, bottom: median dust flux values. Data are divided into the three time stages discussed in the text. Left: Stage III (2.25–0 Ma), centre: Stage II (5.75–3.5 Ma), right: Stage I (11.15–6.9 Ma). Green curves mark the false-alarm level at the 95% confidence level, red curves indicate AR(1) red noise models. Orange lines and numbers indicate the frequencies equivalent to periods (in kyr) of major astronomical cycles (precession, obliquity and eccentricity). Analysis performed and figures created using PAST software[101]. See Supplementary Information for further discussion.



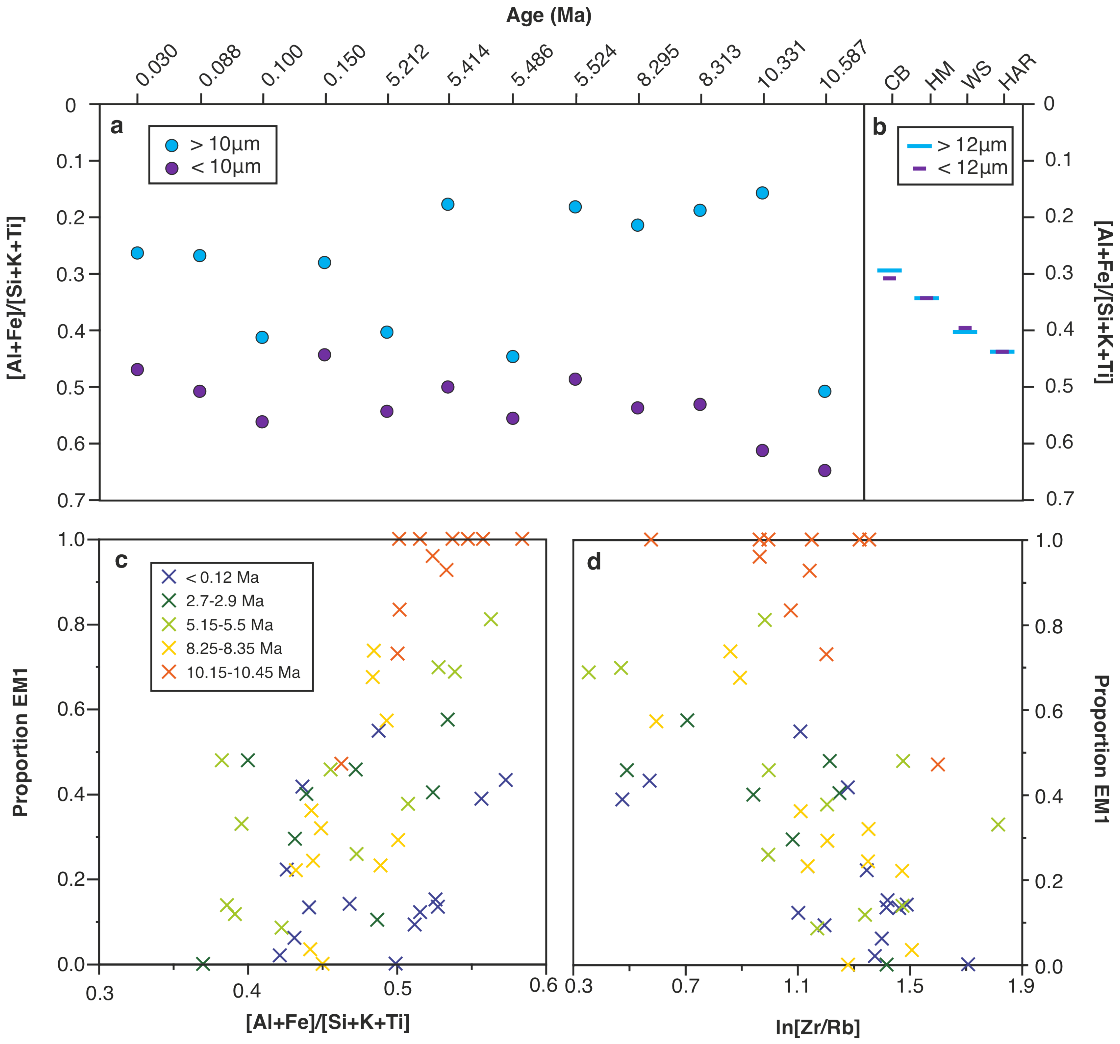
Extended Data Figure 7| Running statistical analysis of Site 659 geochemical data, comparing 1 Myr data bins. **a** Mann-Whitney-Wilcoxon test log(*p*) values to detect shifts in central tendency (see Supplementary Information). Low values indicate extremely low probabilities that the two data bins have the same central tendency. **b** Estimated difference in location between the two data bins divided by the interquartile range of the complete data set, with 95% confidence interval plotted. Note that [Al+Fe] is plotted on an inverted axis. **c** Ansari-Bradley test log(*p*) values to detect shifts in dispersion. Low values indicate extremely low probabilities that the two data bins have the same dispersion. **d** Ratio of scales between the two data bins, with 95% confidence interval plotted. Orange: calibrated [Al+Fe]/[Si+K+Ti], green: ln[Zr/Rb] (with XRF counts <300 removed), red: median dust flux (g cm^{-2} kyr^{-1}). Grey shading indicates intervals of greatest change in the geochemical time series revealed by statistical analyses.



Extended Data Figure 8| Cross-plots of strontium and neodymium isotopic signature of lithic fraction of Site 659 sediments. Data coloured by: **a** the proportion of the lithic fraction attributed to dust by end-member unmixing ([dust]/[dust+riverine]), **b** co-registered ln[Zr/Rb] values. Red marks samples dominated by dust/coarse grains, blue marks samples dominated by riverine inputs/fine grains. **c** Data coloured by age, where stage I (pink) is the oldest (>6.9 Ma) and stage III (blue) is the youngest (<2.25 Ma) and grey indicates samples from the transition between stages II and III (3.5–2.25 Ma). Individual samples are marked by crosses and mean values for each age range shown by circles, with error bars indicating 1 standard deviation.



Extended Data Figure 9| Grain size distributions of the end-members calculated from the lithic fraction of ODP Site 659. Grain size derived end-member 1 (EM1) in blue and grain size derived end-member 2 (EM2) in red compared to: **a** Modern dust samples recorded offshore NW Africa from Stuut et al. (2005) in grey[64]. **b** As (a) but with just the most proximal measurements to Site 659 (M41/1 D4, centered at 19.73°N, 17.91°W) plotted. **c** The grain size end members of Tjallingii et al. (2008) from site GeoB7920-2 (20.75°N, 18.58°W)[12]. Yellow and orange dashed lines indicate the end members attributed to fine and coarse dust respectively, with the riverine end member shown in green. **d** All Site 659 lithogenic grain size distributions (black).



Extended Data Figure 10| Comparisons between grain size and geochemical proxies at Site 659. **a & b** [Al+Fe]/[Si+K+Ti] ratios of the coarse and fine fractions of (**a**) 12 sediment samples from Site 659 sieved at 10µm and analysed by discrete XRF analysis with sample ages are listed along the top and (**b**) desert surface soil and aeolian dusts from four locations in the Sahara-Sahel dust corridor, analysed by inductively coupled plasma mass spectrometry and grouped into <12 µm and >12 µm size fractions, from Castillo et al. (2008)[102]. CB: Chad Basin, HM: Hoggar Massif, WS: Western Sahara, HAR: Harmattan. **c** and **d** The proportion of lithogenic grain size derived end-member 1 (grain size EM1, attributed to fine riverine inputs) plotted against the sediment geochemical ratios **c** [Al+Fe]/[Si+K+Ti] and **d** ln[Zr/Rb]. Data points are coloured by sample age, where the youngest samples are in blue and the oldest in orange.

Supplementary Information

For: Astronomically controlled aridity in the Sahara since at least 11 million years ago

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Supplementary Discussion

Comparison to published dust reconstructions

Reconstructions of the flux of dust from the African continent deposited in ODP Site 659 sediments over the past 5 Myr were originally published by Tiedemann et al. (1989)[1] and Tiedemann et al. (1994)[2]. Their records are based upon the assumption that the proportion of dust in the sediment can be approximated from the non-carbonate fraction (=100–%$CaCO_3$). We find an excellent correlation between the discrete measurements of these earlier studies (which include a range of analytical techniques)[1] and ln[Ca/Fe] values measured by XRF core scanning (Supplementary Figure 4). A Kendall's tau-b correlation test between the dust % estimates of Tiedemann et al. (1994)[2] and those presented here reveals a positive, extremely significant correlation ($\tau = 0.669$, *p*-value <0.001), with a comparison between published dust % to ln[Ca/Fe] revealing an even stronger (but negative) correlation ($\tau = -0.711$, p-value <0.001). These results show that our ln[Ca/Fe] data provide reliable down core records of terrigenous sediment content at ODP Site 659. We note, that these datasets, in common with many palaeoclimate records, likely show autocorrelation (where there is a degree of similarity between a time series and a lagged version of itself). This acts to elevate the calculated correlation coefficients between data series. We therefore also show coherency spectra, calculated using the REDFIT-X program of Ólafsdóttir et al. (2016)[3]. This approach uses a Lomb-Scargle Fourier transform to perform cross-spectral analyses between unevenly spaced data sets. Monte Carlo simulations that mimic key properties of the datasets (e.g. autocorrelation and time spacing) are used to estimate the false-alarm level for the calculated coherency. These spectra are shown in Extended Data Figure 2. Significant coherency between the dust % estimates of Tiedemann et al. (1994) and both our ln[Ca/Fe] and dust % estimates across almost all frequencies supports the strong agreement among the different techniques (Extended Data Figure 2a,b).

Recent work has shown that, in addition to dust inputs, fine-grained riverine sediment is a likely contributor to Site 659 and the nearby continental margin[4,5]. We therefore partitioned the bulk terrigenous sediment component into two separate components (classified as riverine and aeolian) by applying an unmixing approach (Methods). Our absolute dust fluxes are therefore lower than those of Tiedemann et al. (1994)[2] (Supplementary Figure 4), which were calculated by assuming that all terrigenous material at Site 659 was wind-blown in origin. Nevertheless, for the last 5 Myrs, where there are detailed records from Tiedemann et al. (1994)[2], the down core trends in the data yielded by the two different approaches are generally very consistent over both secular and astronomical timescales.

For sediments younger than about 200 to 300 kyr, thorium-normalization of sediment accumulation rates has been used to provide age control independent of sediment focusing and dissolution along the NW African margin[6-10]. To assess whether these processes likely exerted a major control over our dust flux reconstructions we compared our calculated fluxes to those from site MD03-2705[9] (18°05′N, 21°09′W). Because dust flux estimates were calculated at different age points between the two sites, we calculated generalized additive models (GAMs)[11] using R[12] to create a smoothed fit to our datasets and resampled this at ages corresponding to the ^{230}Th-normalised data points (Extended Data Figure 3). This approach provides a good representation of the original dataset, except for the most recent 20 kyr, where GAMs struggle to capture the high frequency, high magnitude variability seen at both Site 659 and MD03-2705. The degree of correlation between the two dust flux reconstructions was then assessed using a Kendall's tau-b correlation test. Despite uncertainty introduced by the GAM approximations and chronological uncertainties between the two sites, we find a positive, strongly significant correlation ($\tau = 0.298$, p-value <0.001) between our dust fluxes at Site 659 and those calculated by ^{230}Th-normalisation at proximal site MD03-2705[9] (Extended Data Figure 3). We also performed cross-spectral analysis between the two datasets using REDFIT-X[3] (where no resampling is required). The results of these analyses indicate that there is largely significant coherency between data sets across the frequency range 0.025–0.23 cycles/kyr (Extended Data Figure 2c), equivalent to periodicities of 40–4 kyr. These results suggest that our dust flux reconstructions are representative of aeolian transport from the continent and not major sediment redistribution in the ocean or rapid pulses of dissolution, at least over the past 240 kyr where ^{230}Th-normalised dust flux estimates exist.

Comparison to established XRF ratios and other hydroclimate proxies

Our discussion focuses on two geochemical ratios ([Al+Fe]/[Si+K+Ti] and ln[Zr/Rb]) provided by our XRF work but our interpretations gain further support from three other elemental ratios that have also been shown to closely track climatic gradients along the African coastline today. Ti/Al is proposed as a proxy for aeolian versus fluvial input and Fe/K and Al/Si are suggested as tracers of terrestrial climatic zones[13], although Al/Si can also be influenced by high siliceous microfossil concentrations at times of intense upwelling[14]. All three of these ratios agree very closely with one another and with [Al+Fe]/[Si+K+Ti] over astronomical timescales throughout our records (Supplementary Figures 5 and 6). Dark sediment layers consistently record high ln[Ti/Al] values and low ln[Fe/K] and ln[Al/Si] values, indicating a dominance of aeolian over riverine sediment and a decrease in the intensity of chemical weathering, a trend documented with increasing aridity associated with increasing distance from the equator today[13]. These findings strongly support our interpretation that the dark sediment bands represent dust pulses associated with increased aridity throughout the last 11 Myr.

The different elemental ratios also show clear similarity over multi-million year timescales. The onset of more extreme wet events between 7 and 6 Myr can be seen in all ratios, as can a shift towards more arid conditions over the past ~3 Myr. A decrease in ln[Al/Si] through the Pliocene and Pleistocene is more pronounced than the other ratios, and, given the very low concentrations of siliceous microfossils recorded in the Site 659 sediments[15], may indicate an increase in the proportion of silica-rich terrigenous material reaching the site, such as pale quartz grains from coastal sources[16].

We also compare our [Al+Fe]/[Si+K+Ti] and ln[Zr/Rb] data to an independent hydroclimate proxy: the hydrogen isotopic (δD) signature of *n*-alkanes (Extended Data Figure 1). These compounds are found as leaf waxes in terrestrial plants, with δD values at Site 659 suggested to largely depend upon the amount of rainfall, although differences in plant physiology (e.g. photosynthetic pathway, rooting depth and leaf anatomy) and climate (e.g. moisture source region, evaporation) can also influence plant wax δD signatures[17]. Extended Data Figure 1 illustrates the strong agreement between C_{31} *n*-alkane δD values and our XRF-based hydroclimate proxies during several snapshots of the Pliocene and Pleistocene (where δD data are available)[18,19].

We also test the similarity between the datasets statistically. Our data and the reported δD values are from the same site but different sample sets were used so we apply GAMs to create a smoothed fit to our [Al+Fe]/[Si+K+Ti] record and resample it at the matching depths to the δD data (Extended Data Figure 1d-f). Separate fits were made for 0.14–0 Ma, 3.27–3 Ma, 3.63–3.34 Ma, 4.83–4.61 Ma and 4.99–4.83 Ma to ensure that the GAMs captured astronomical-scale variability. We combined the data from all of the time windows to perform a Kendall's tau-b correlation test with the full δD dataset, which revealed a negative, strongly significant correlation with [Al+Fe]/[Si+K+Ti] ($\tau = -0.218$, p-value <0.001). Note that a negative correlation indicates agreement between the proxies, with higher [Al+Fe]/[Si+K+Ti] values but more negative δD values indicating a wetter climate. The strength of the relationship between our XRF data and the δD results (Extended Data Figure 1a-c) support the use of [Al+Fe]/[Si+K+Ti] to reconstruct past changes in continental hydroclimate in these sediments.

We also performed cross-spectral analysis to assess how the coherency of [Al+Fe]/[Si+K+Ti] and δD varies with frequency (Extended Data Figure 2d-f). These analyses were carried out for three separate time slices due to the discontinuous nature of available high-resolution δD data. We find the strongest coherency occurs at frequencies of ~0.03–0.05 cycles/kyr, implying that these records are coherent over precessional timescales, both responding to insolation-forced variability in hydroclimate. Outside this range, the coherency is lower. This result may indicate the influence of secondary processes acting on one or both proxies over different timescales.

Geochemical differences between light and dark layers

Colour data were generated from the central portion of ODP core photograph images[15] along our splice using Code for Ocean Drilling Database macros[20], with an average sample spacing of ~2mm. Lightness values of >80 or more than 3 standard deviations above or below a 4 cm rolling mean value were excluded to filter out artefacts such as cracks and core gaps from the dataset. Lightness data were then smoothed by calculating the mean value over a 4 cm rolling window.

To identify "light" and "dark" sediment layers, we filtered out longer-term variability in the lightness data by calculating residuals from a 5 m rolling mean. The residual dataset was then resampled to match the depths corresponding to XRF data using simple linear interpolation. "Light" layers were defined as those with residual lightness values greater than the 60% percentile of the rolling 5 m window, while dark layers were defined as those with lightness values below the 40% percentile.

Extended Data Figure 4 illustrates the geochemical differences between sedimentary layers classified as "light" and "dark" in the Site 659 XRF datasets. Mann-Whitney-Wilcoxon tests were used to identify whether there were significant geochemical differences between the light and dark layers for each of our three time intervals. Extremely low p-values (<0.001) are documented for both [Al+Fe]/[Si+K+Ti] and ln[Zr/Rb] in each of the three time windows, which shows that there are clear, highly significant geochemical differences between light and dark sediment layers at Site 659.

Identification of change points in time series

To identify the intervals of greatest change in our geochemical time series, we applied running Mann-Whitney-Wilcoxon (also known as Wilcoxon rank sum) and Ansari-Bradley statistical tests, following the approach of Trauth et al. (2009)[21]. We test whether samples taken from 1 Myr ranges either side of a selected age come from distributions with the same properties, or whether there were significant differences between them. We used 1 Myr bins to smooth out the influence of shorter frequency astronomical variability and highlight secular changes in the datasets.

[Al+Fe]/[Si+K+Ti], ln[Zr/Rb] and median dust flux data sets were each resampled using simple linear interpolation to give an average data resolution of 2 kyr. A Mann-Whitney-Wilcoxon test was applied to test the null hypothesis that samples from the million years preceding and postdating the chosen date were selected from identical continuous distributions with equal medians.

The Ansari-Bradley test determines whether two samples come from continuous distributions with the same median and shape but different dispersions. As the results of the Mann-Whitney-Wilcoxon test indicate that there are significant shifts in the median values of all three of our proxy data sets between different time windows, we calculate the difference of each data point from the whole-core mean and then adjust these residuals by the difference in location between the two time windows (the median of the difference between a sample from each of the two distributions). Due to the size of the datasets, a normal approximation was used when calculating the *p*-values.

The results of the running Mann-Whitney-Wilcoxon and Ansari-Bradley tests on our three proxy datasets reveal two major intervals of change in our time series; 6.9–5.75 Ma and 3.5–2.25 Ma (Extended Data Figure 7). The first interval is marked by a major increase in the variability of all three proxy records from 6.9–6.3 Ma, accompanied by an increase in the central tendency of [Al+Fe]/[Si+K+Ti] values. This is followed by a decrease in ln[Zr/Rb] values (6.2–5.75 Ma).

During the second key interval of change, a decrease in the central tendency of [Al+Fe]/[Si+K+Ti] (centred around 3.2–3.1 Ma) again leads change in the ln[Zr/Rb] data (which shows an increase centred around 2.8–2.25 Ma), although this time there is also an increase in the mean dust flux values approximately synchronous with the change in ln[Zr/Rb]. There is a shift to greater dispersion in the dust fluxes centred around 2.7 Ma, but no concurrent change in the amplitude of variability of [Al+Fe]/[Si+K+Ti] or ln[Zr/Rb].

Attribution of sedimentological end-members

Our two lithogenic grain size end-members identified at Site 659 have clearly distinct grain size distributions. End-member EM1 is by far the finer of the two, with modal grain sizes of 0.7–4 μm. End-member EM2 has a sharp, unimodal distribution dominated by grains of 10–40 μm diameter.

There is remarkable agreement between the grain size distributions of Site 659 lithogenic end-member 2 (EM2) and dust samples taken offshore NW Africa[22], particularly those sampled closest to Site 659 (Extended Data Figure 9). EM2 is also similar to the fine dust end-member identified at NW African site GeoB7920-2 by Tjallingii et al. (2008)[4] (Extended Data Figure 9). We therefore attribute EM2 to windblown dust exported from the African continent.

The origin of EM1 is less immediately obvious. Distal dusts transported long distances from their sources are suggested to have volume median grain sizes of ca. 2–7 μm[23-26], therefore, it is possible that EM1 at Site 659 also represents a dust component, albeit much finer than EM2. However, Site 659 end-member 1 (EM1) contains a much higher proportion of grains <2 μm than any of the atmospheric dust samples recorded offshore northwestern Africa by Stuut et al. (2005)[22] (Extended Data Figure 9a), and is finer than either samples dominated by either wet or dry deposition captured by sediment traps offshore Cape Blanc[27]. EM1 is also finer than modern African dust captured across the Atlantic throughout the year and more distal African-sourced dust in Puerto Rico[23,28,29]. These observations strongly suggest that EM1 is unlikely to represent a pure dust component, at least under wind conditions similar to modern.

We note that EM1 has a very strong similarity to the grain size distribution of the fine lithogenic end-members identified by both Holz et al. (2004)[30] and Tjallingii et al (2008)[4], which are attributed to distal rain-out of fine particulates from riverine sources. Networks of river channels, active during past intervals of humidity have been identified across North Africa, many of which drain into the Atlantic[5,31], with submarine channels transporting this material into the deep ocean[32,33]. A distal riverine interpretation for fine-grained sediments at Site 659 is supported by high proportions of the fine end-member in core top sediments offshore Morocco which receive sediment from the Souss River[30] and the fact that highest proportions of this fine end-member occur at times where enhanced rainfall is well evidenced in North Africa, such as African Humid Period 1[4,34], although increased fluxes of fine sediments are not consistently observed at all sites on the northwest African margin at this time[8]. The elevated location of Site 659 on the Cape Verde Plateau means that hypopycnal transport would likely be required for material of riverine origin to reach the site. EM1 at Site 659 is slightly finer than the riverine end-member identified at site GeoB7920-2[4] (Extended Data Figure 9c), which is consistent with the greater distance from the continent of Site 659 compared to GeoB7920-2 and the requirement for sediment to be suspended within the water column to reach Site 659 due to its elevated position on the Cape Verde Rise (Supplementary Figure 1).

A third possible contributor to EM1 is resuspension of sediments from the continental shelf. Although shelf sediments are dominated by grains much coarser than the aeolian inputs, potentially representing relict dune fields[35], fine grained components are preferentially transported in nepheloid layers[36]. Nepheloid layer activity is currently very low around Cape Verde[37,38], however, a contribution of these processes to Site 659 cannot be ruled out.

To further explore the origin of EM1, we test for geochemical differences between the fine and coarse components of the sediments. 12 samples with a range of ages and lithologies were wet sieved at 10 μm (close to the boundary between EM1 and EM2 dominance) to separate out the coarse and finer fractions. Samples were then oven dried and homogenized, then mixed with di-lithium tetraborate and fused to create beads. These were analysed by X-ray fluorescence using a Phillip MagiX Pro WD-XRF at the University of Southampton Waterfront Campus, NOCS, calibrated using international rock standards prepared in the same way as our samples.

All 12 of our samples show a clear offset in [Al+Fe]/[Si+K+Ti] between the >10 μm and <10 μm size fractions (Extended Data Figure 10a). The <10 μm [Al+Fe]/[Si+K+Ti] values are 0.11–0.46 greater than the <10 μm fraction, which means that the fine grained components plot much closer to the value of suspended sediments from the Senegal River than the coarse fractions do. The largest size fraction differences are generally recorded in the Miocene aged samples from depths with high terrigenous contents (indicated by high ln[Fe/Ca] values and a dark colour). In addition, there is a strong correlation between the relative proportions of grain-size end-members and our geochemical ratios for past dust inputs (Extended Data Figure 10c,d). High proportions of EM1 are associated with both high [Al+Fe]/[Si+K+Ti] values (similar to modern riverine inputs) and low ln[Zr/Rb] values which are commonly interpreted as indicating a dominance of fine-grained clays over coarse dust[4,39]. These results are strongly supportive of a distal riverine contribution to EM1.

We note that differences in major element compositions might also be expected between coarse and fine fractions of a purely aeolian sample due to the susceptibility of different minerals with different chemical compositions to be weathered into finer particles. However, samples of dusts and dust source sediments from across the Sahara-Sahel dust corridor show negligible offsets in [Al+Fe]/[Si+K+Ti] between size fractions (Extended Data Figure 10b)[40]. Therefore, we suggest that the differences we record in major element signatures between the <10 μm and >10 μm size fractions reflect inputs from multiple sources rather than size-dependent fractionation from a single source.

The higher [Al+Fe]/[Si+K+Ti] values of the <10 μm sediment component (Extended Data Figure 10) mean that our geochemical unmixing approach acts to remove this very fine component. Therefore, even if EM1 is dominantly derived from resuspended shelf material rather than distal riverine material, the geochemical unmixing approach will still act to remove its contribution from the total lithogenic component to produce our dust flux estimates. If EM1 contains a component of fine-grained dust, then our dust flux estimates are likely to be underestimates.

In conclusion, a dominantly distal riverine origin for EM1 is indicated by: (i) grain size distribution analyses showing that EM1 is very similar to the fine end-member previously identified at higher concentrations during humid time intervals[4,34] and at latitudes to the north of the Sahara where precipitation is higher[30] (ii) EM1 is much finer than either proximal or distal North African dust (including samples from as far as the Caribbean)[22,23,29,41], (iii) large geochemical differences between the coarse and fine fractions of Site 659 sediments are too great to be explained by size-dependent fractionation from a single source, and (iv) the current absence of major nepheloid layer activity around Cape Verde[37,38].

Effects of grain size on radiogenic isotope signatures

A grain size effect on Sr isotope signatures of sedimentary lithics has previously been documented including in African dust, with fine-grained samples having higher $^{87}Sr/^{86}Sr$ values, although a similar effect in Nd isotopes is not commonly observed[42-44]. We find no strong relationship between the radiogenic isotopic signature and either the proportion of dust or the grain size of the sediment as approximated by ln[Zr/Rb] (Extended Data Figure 8a,b), although we note a tendency for samples with the most radiogenic Sr and unradiogenic Nd isotope signatures to have low ln[Zr/Rb] values and be dominated by riverine inputs rather than dust (according to the results of our geochemical end-member unmixing), with the inverse also true, particularly in ln[Zr/Rb]. Radiogenic Sr values may be attributed to enhanced chemical weathering under humid climates resulting in a higher proportion of fine clay minerals and micas reaching Site 659[42]. The covariation with unradiogenic ε_{Nd} values suggest there is also a shift in provenance, potentially attributable to transport of an increased proportion of proximal material sourced from the West African Craton transported by rivers compared to aeolian inputs, which may contain a greater contribution from the interior of the continent (or potentially Morocco, although the reported Sr isotope signatures[45] suggest this scenario is less likely).

Temporal variability in radiogenic isotope signatures

The radiogenic isotope signatures of the lithic fraction of the Site 659 sediments hint at subtle differences with sediment age, but we record no clear temporal shifts in the provenance of lithogenic material through the last 11 Myr to suggest major changes in the source regions of terrigenous sediment. The oldest samples (stage I) generally show more non-radiogenic isotope values in $^{87}Sr/^{86}Sr$ (Extended Data Figure 8c), which means that these Miocene-aged samples plot slightly closer to the Central source area than to the Western region, as defined by the currently available data[46] (Figure 4). Therefore, we infer that the strong signal of dust inputs during stage I cannot be attributed to input from coastal sand dunes (which have previously been documented back to the Early Miocene and potentially much earlier[47,48]) and instead indicate the presence of strong dust generating regions in the continental interior back to at least the Late Miocene.

Fidelity of an astronomically-tuned age model at Site 659

Pronounced lithological and geochemical variability is well documented at Site 659, particularly over the younger intervals, and has been linked to astronomical forcing[2,19,49-51] (e.g. Figure 3, Supplementary Figure 6). These observations therefore provide an opportunity to capitalize on the potential to gain a high-fidelity age model for Site 659 by tuning the clear lithological cycles to an astronomical signal, as with many records of Mediterranean climate and low-latitude continental margins where a similar tuning approach has been used for chronology[52-56]. However, this approach requires care to ensure that age is assigned to our environmental records as independently as possible to guard against circularity. In our final age model, we used the core images and the ratio ln[Ca/Fe] to tune to insolation and [Al+Fe]/[Si+K+Ti] and ln[Zr/Rb] as our main records of environmental change.

To assess the extent to which astronomical power is present throughout the Site 659 records, we first analysed the frequencies present in our data using wavelet analysis[57] with an age model was based solely on bio- and magnetostratigraphic datums[15,58] and no tuning applied to the records (Extended Data Figure 5). The resulting continuous wavelet power spectrum show extremely similar patterns of variability on astronomical periodicities to our final, astronomically tuned record in both ln[Ca/Fe] and [Al+Fe]/[Si+K+Ti] across the full 11 Myr datasets (Extended Data Figure 5). This result indicates that the astronomical periodicities identified in the lithology and geochemistry of Site 659 sediments are genuine and have not been artificially tuned into the data. We also note that significant power is often detected concurrently at two or more frequencies, with the ratios between these frequencies matching those of major astronomical cycles (Extended Data Figures 5 and 6). This result would be very difficult to tune into our records artificially, providing further evidence for the lithological and geochemical variability at Site 659 being astronomically-paced.

Changing astronomical pacing of hydroclimate variability in North African over the past 11 Myr

To assess how the dominant frequencies of variability change throughout our >11 Myr records, we performed spectral analysis on our ln[Ca/Fe], [Al+Fe]/[Si+K+Ti] and dust flux data using REDFIT[59] to identify significant periodicities in our unevenly spaced data series (Extended Data Figure 6). Analyses were carried out using the PAST software[60]

REDFIT analysis reveals a strong signature of precession throughout the last 11 Myr in ln[Ca/Fe], [Al+Fe]/[Si+K+Ti] and dust flux data from Site 659 (Extended Data Figure 6), with obliquity also present in all three time intervals examined (stages I to III). Significant eccentricity is only recorded during stage I (>6.9 Ma), rather than the youngest 800 kyr when strong 100 kyr glacial cycles are recorded in benthic oxygen isotope records[61]. The signatures of obliquity and precession are seen in all three stages of our record, but are generally weakest in the oldest part of the record (stage I), likely, at least in part, because of the stronger smoothing effects of bioturbation where sedimentation rates are lower (Supplementary Figure 2).

The relative importance of high and low latitude forcing in driving African hydroclimate, particularly during the Quaternary Period, is debated[2,9,62-67]. The strong presence of precessional frequencies (23 and 19 kyr) in our data (Extended Data Figure 6) supports a major role for insolation forcing of the West African monsoon. Obliquity (41 kyr) is also strongly present, which may indicate that the ice volume at high latitudes also exerts a major control on North African hydroclimate, perhaps by influencing wind strength. However, obliquity can also influence low latitude hydroclimate directly through modulation of the cross-equatorial insolation gradient[68]. The absence of an obvious strengthening of the obliquity signature associated with increasing amplitude of glacial-interglacial cycles in the Late Pliocene/early Pleistocene[61] in our hydroclimate proxy [Al+Fe]/[Si+K+Ti] (e.g. Extended Data Figure 5) supports this interpretation. The signal of obliquity in the latest Pliocene/Pleistocene appears slightly stronger in the ln[Ca/Fe] and dust flux records than [Al+Fe]/[Si+K+Ti] which may indicate the influence of glacial-interglacial shifts in overturning circulation bringing more corrosive bottom waters to the site driving carbonate dissolution[9] and/or higher latitude forcing exerting a stronger control over wind strength (and hence dust transport) than continental hydroclimate.

Interpreting plant wax history at Site 659

Long-chain *n*-alkanes originating from the epicuticular leaf waxes of terrestrial plants can be removed from the leaf surface by the action of wind or rain and transported to the ocean via both dust and rivers[69-71]. A strong correlation between concentrations of *n*-alkanes and dust percentages at Site 659 suggests that transport by wind is the dominant process at this location[2,19]. The source regions of plant waxes are likely very similar to those of the terrigenous material reaching Site 659, however, given the very low net primary production across much of North Africa, the plant wax signature may be biased to areas with higher biomass densities, particularly during arid time intervals.

Previous studies suggest that Site 659 records plant waxes dominantly sourced from the northern Sahel, with biomarker and pollen records suggesting a minor additional input of terrestrial material transported by the northeast trade winds from more northerly latitudes, evidenced during times of aridity and increased trade wind strength such as the Last Glacial Period, but not during the warmer and wetter Pliocene[18,19,72,73]. The Sahel is currently dominated by C_4 grasslands, while the vegetation in northernmost Africa is instead dominated by C_3 plants[74]. Therefore, the northwest African margin currently receives a mixture of plant waxes from these two sources. Several studies[72,75] estimate approximately equal proportions of inputs from C_4 and C_3 plants to recent and Late Quaternary aged marine sediments around the latitude of Site 659, however other, more recent studies point to a much higher contribution from C_4 plants[18,76].

Combined evidence from plant wax carbon isotopes and pollen indicates C_4 savanna grasslands directly replaced savannas dominated by C_3 grasses in both eastern and southern Africa as C_4 vegetation expanded through the Neogene[77,78]. Evidence of Poaceae (grasses) in the Niger delta as far back as the early Miocene[79] suggests a similar sequence of events unfolded in the Sahel, with a gradational increase in plant wax $\delta^{13}C$ of ca. 10 ‰ at Site 659 indicating an expansion of C_4 vegetation starting approximately 10 Myr ago[80]. Superimposed on this shift (at least during the Pliocene and Late Quaternary where high-resolution data exist) is variability in $\delta^{13}C$ driven by precession, obliquity and eccentricity[19]. These cycles have a typical magnitude of 0.5–2.5 ‰, much smaller than the multi-million year trend in plant wax $\delta^{13}C$ (Figure 2, Supplementary Figure 3).

We note that the relatively muted signature of astronomically-forced variability recorded at Site 659 is not observed at all locations across North Africa. Some localities record variability in plant wax $\delta^{13}C$ of >8 ‰ over astronomical timescales[81-86], most commonly records from lakes in East Africa where the topography can lead to diverse ecosystems in closer proximity to one another than further west. Lacustrine sediments also typically record larger variability in plant wax $\delta^{13}C$ than distal marine sediment cores[87] because lake archives document more proximal vegetation sources, can be influenced by local tectonics and hydrography particularly in tectonically active regions such as East Africa, and may also be biased by microbially-derived *n*-alkanes[87]. Marine records integrate source material from across a large region and are less sensitive to subtle vegetation shifts than lake records although they can be influenced by transport processes[19,87].

A larger range of variability in *n*-alkane $\delta^{13}C$ in the Late Quaternary than Site 659 is also recorded at northwest African margin sites GeoB9508-5 (15.5°N, -21.2 to -26.5 ‰, ~65–100 % C_4)[76] and GeoB9528-3 (9.2°N, -21.8 to -28 ‰, ~55–100 % C_4)[88] (21.0°N, -23 to -25.7 ‰, ~70–90% C_4). Both GeoB9508-5 and GeoB9528-3 lie further south and closer to the continent than Site 659. Therefore, a scarcity of sources of waxes carrying very low carbon isotopic signatures in the Sahara due to a regional dominance of C_4 vegetation could be responsible for low amplitude variability in $\delta^{13}C$ at Site 659. That said, we note that the amplitude of astronomically-forced variability in *n*-alkane $\delta^{13}C$ at both GeoB9508-5 and GeoB9528-3 is only around half the magnitude of the multi-million year shift associated with C_4 grassland expansion. We also observe that the magnitude of astronomically-forced variability in $\delta^{13}C$ at Site 659 appears slightly smaller in the Pliocene than in the Last Glacial Cycle, despite lower $\delta^{13}C$ values indicating a greater proportion of C_3 plants in the older time period (Figure 2, Supplementary Figure 3). The *n*-alkane $\delta^{13}C$ data from the Late Miocene are not of sufficient resolution to assess the magnitude of astronomically-forced variability, however we do not observe an obvious relationship between $\delta^{13}C$ and our geochemical ratios to suggest that there was strong aridity-driven cyclicity at a time when isotopic signatures indicate significant inputs from both C_3 and C_4 plants.

In summary, we conclude that it is unlikely that the muted astronomically-forced variability in $\delta^{13}C$ recorded in both the Pliocene and Quaternary at Site 659 (compared to the high amplitude astronomically-forced changes in hydroclimate) can simply be explained by the absence of a C_3 plants in the regions of Northern Africa sampled by our records.

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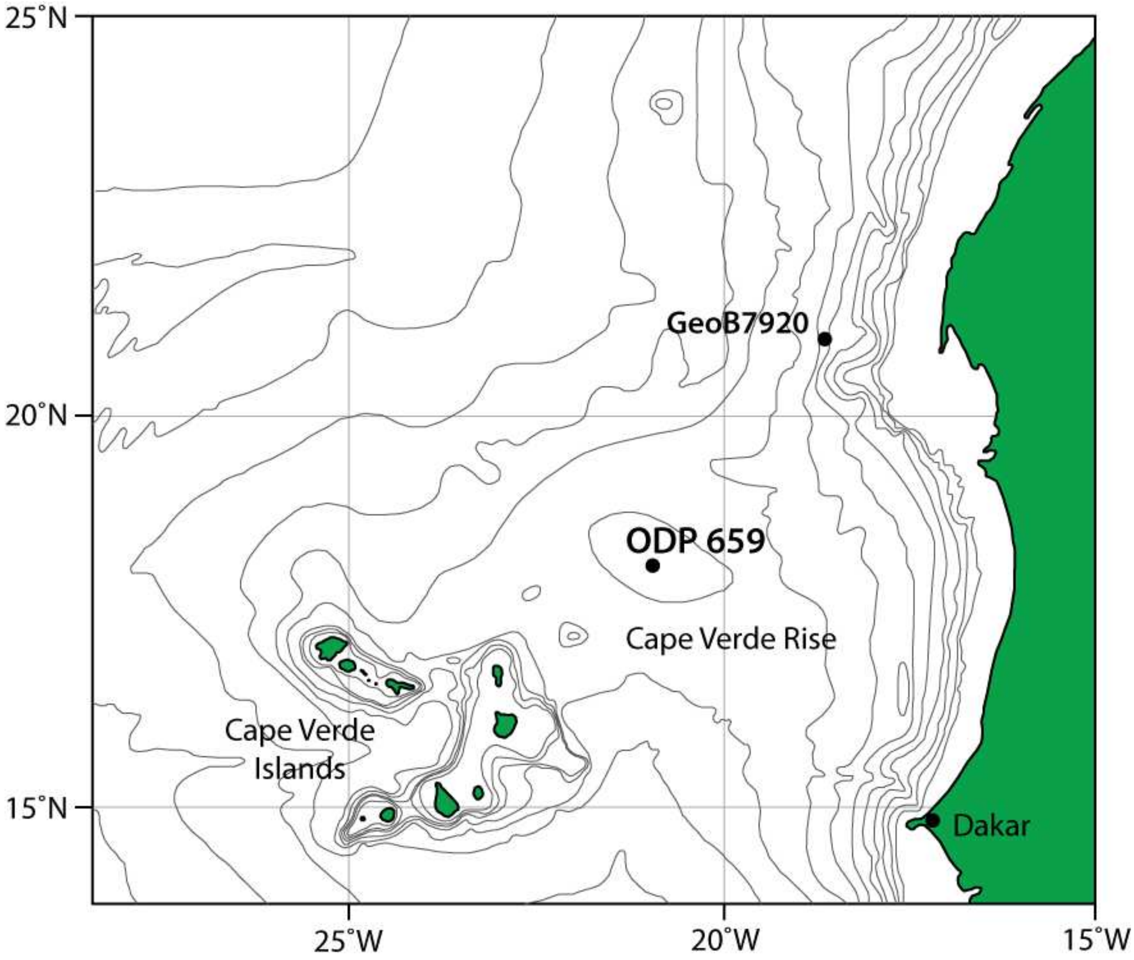
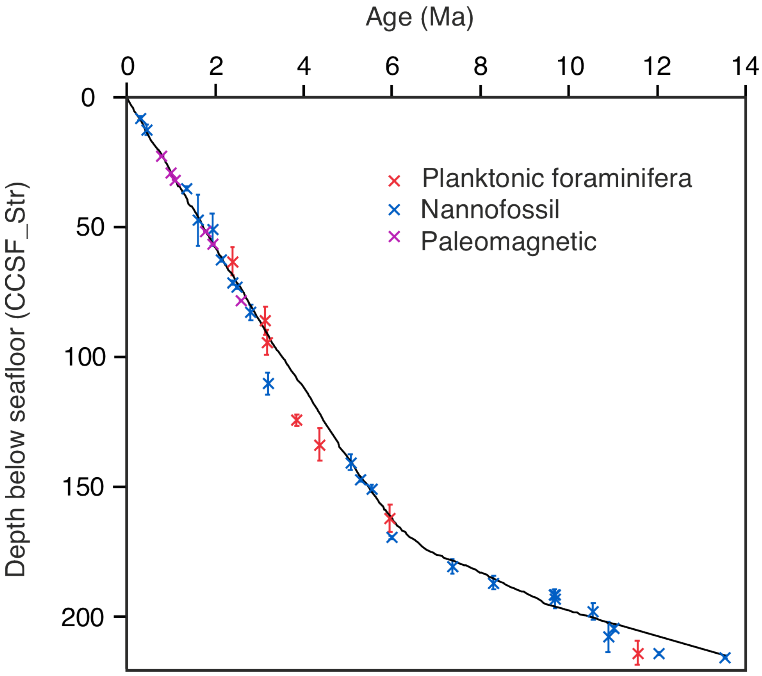
Badger et al. (2019)[90]
Bartoli et al. (2011)[91]
Bolton et al. (2016)[92]
Chalk et al. (2017)[93]
de la Vega et al. (2020)[94]
Dyez et al. (2018)[95]
Hönisch et al. (2009)[96]
Martinez-Boti et al. (2015)[97]
Pagani et al. (2005)[98]
Pagani et al. (2010)[99]
Seki et al. (2010)[100]
Sosdian et al. (2018)[101]
Stap et al. (2016)[102]
Super et al. (2018)[103]
Tanner et al. (2020)[104]
Zhang et al. (2013)[105]
Zhang et al. (2017)[106]
Zhang et al. (2019)[107]

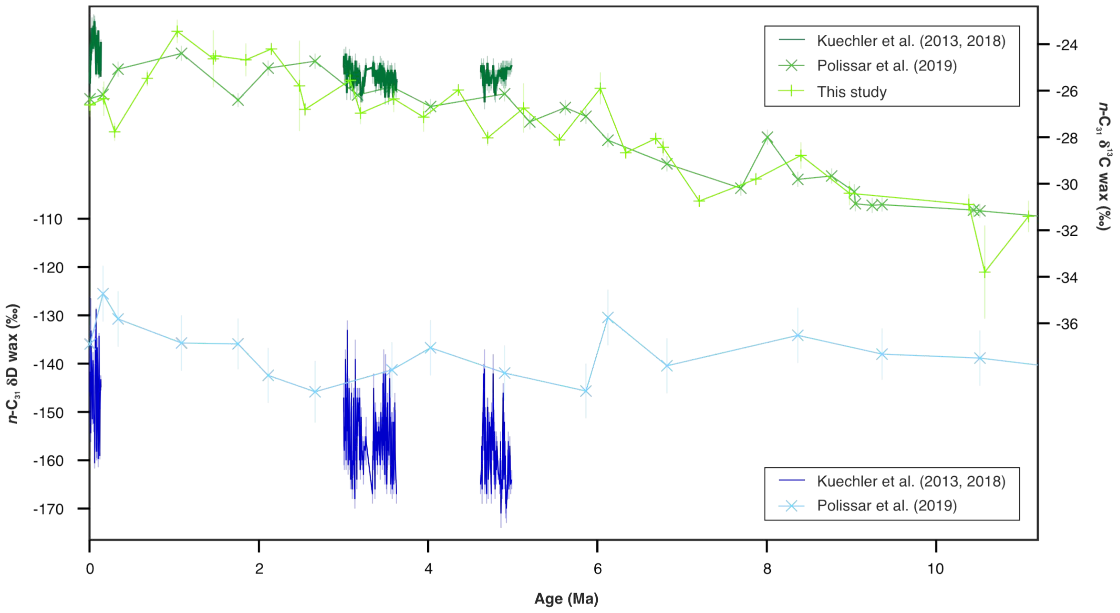
Supplementary Figures



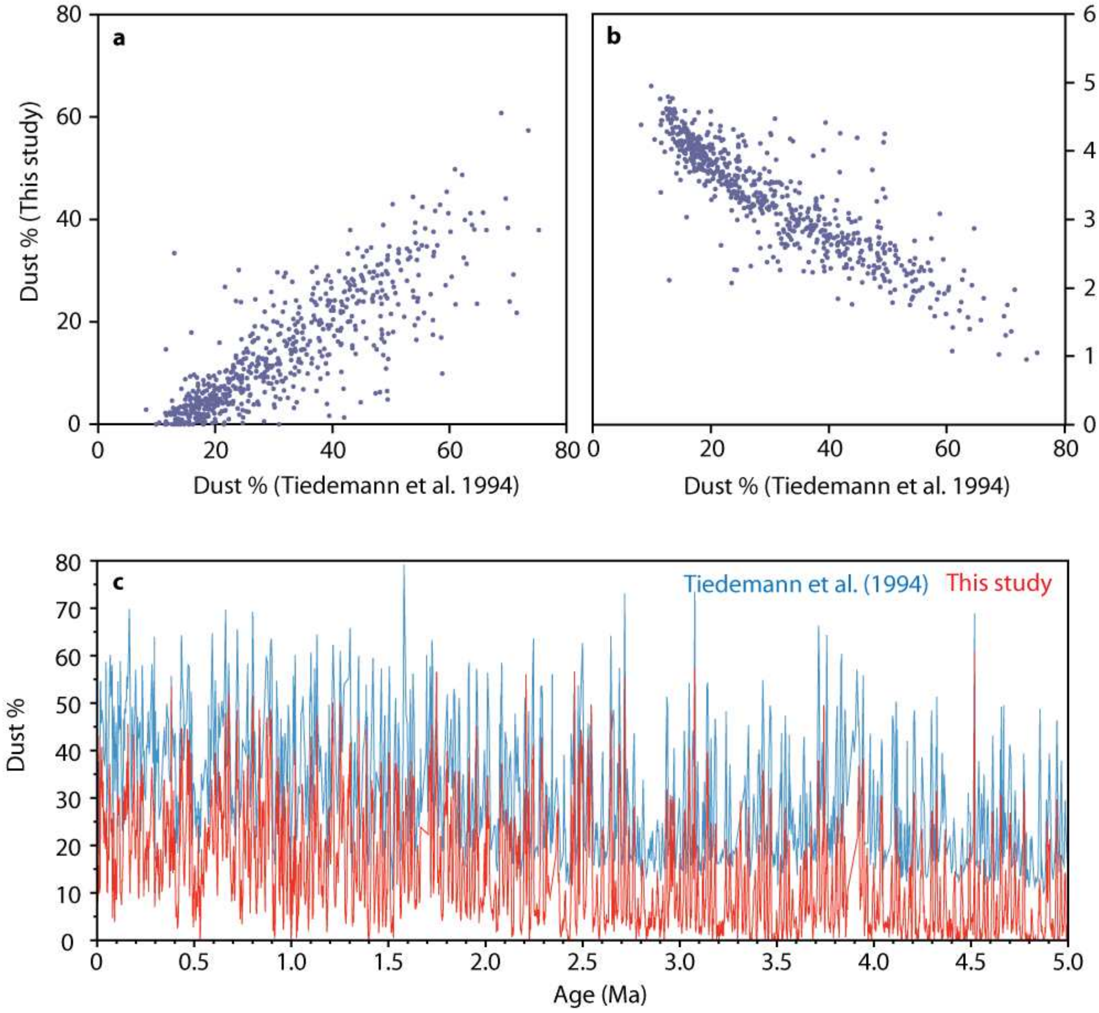
Supplementary Figure 1| Map indicating the location of ODP Site 659 and nearby site GeoB7920, offshore NW Africa. Site locations marked by black circles, adapted from Ruddiman et al. (1989)[15].



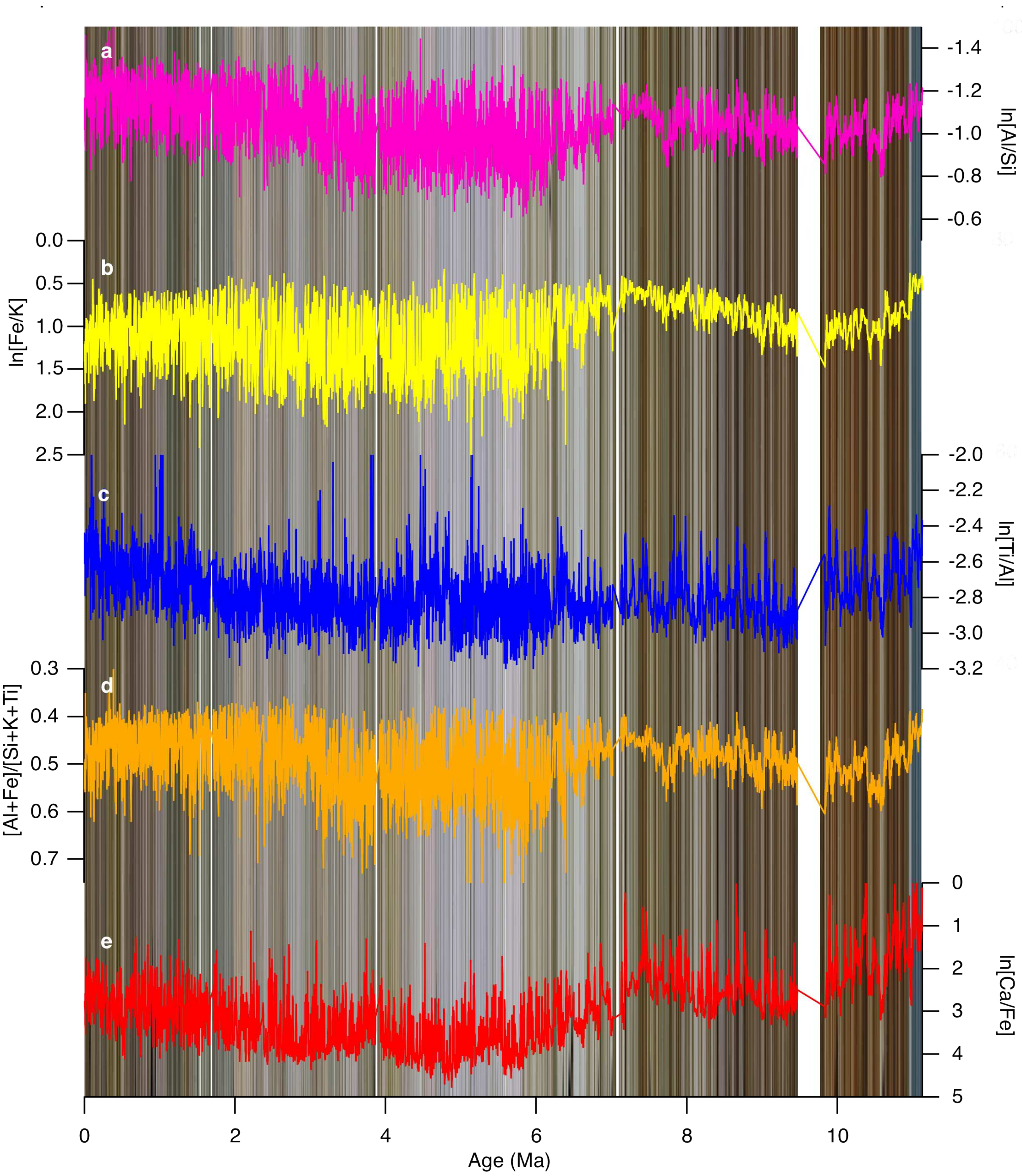
Supplementary Figure 2| Age depth plot for ODP Site 659. Age model created by benthic oxygen isotope correlation and orbital tuning used in this study (black line) compared to the original datums[58,108] (J. Backman pers. comm.), with depths converted onto the new splice and out-of-splice sections stretched to correlate to the splice where necessary using XRF data, bulk properties and core images. Biostratigraphic and magnetostratigraphic ages updated following Raffi et al. (2006)[109], Wade et al. (2011)[110] & Ogg (2012)[111]. Red crosses: planktonic foraminiferal datums, blue: nannofossil datums, purple: paleomagnetic reversals.



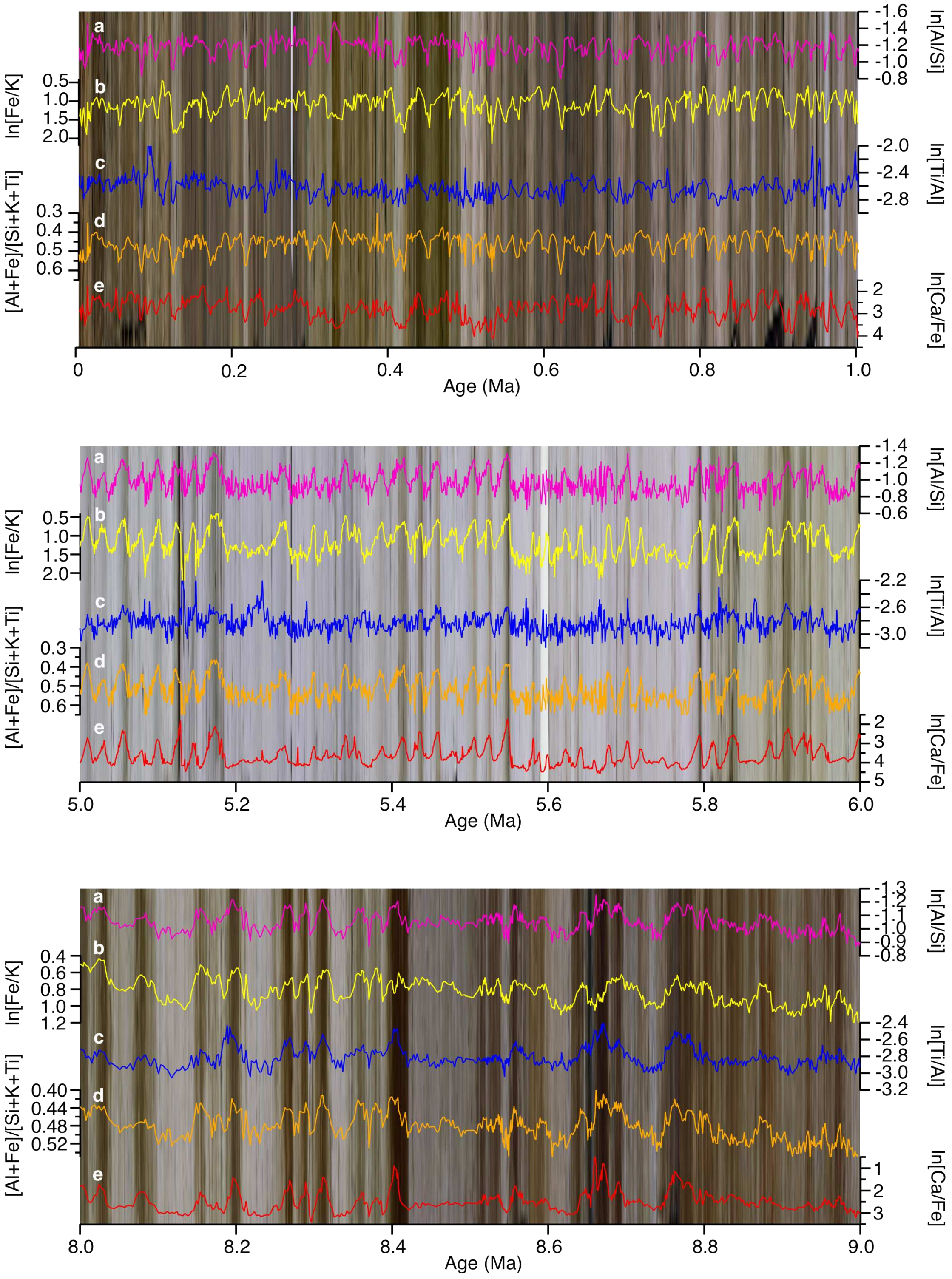
Supplementary Figure 3| Comparison of astronomically-forced variability and multi-million year trends in plant wax isotopes at Site 659. Top: δ^{13}C of C_{31} *n*-alkanes, data from Kuechler et al. (2013, 2018, line only)[18,19], Polissar et al (2019, crosses)[80] and this study (plusses). Bottom: δD of C_{31} *n*-alkanes, data from Kuechler et al. (2013, 2018, line only)[18,19] and Polissar et al (2019, crosses)[80]. Error bars indicate 1 s.e.m in the data of Polissar et al. (2019) and 1 sd in the data from both Kuechler et al. (2013, 2018) and this study.



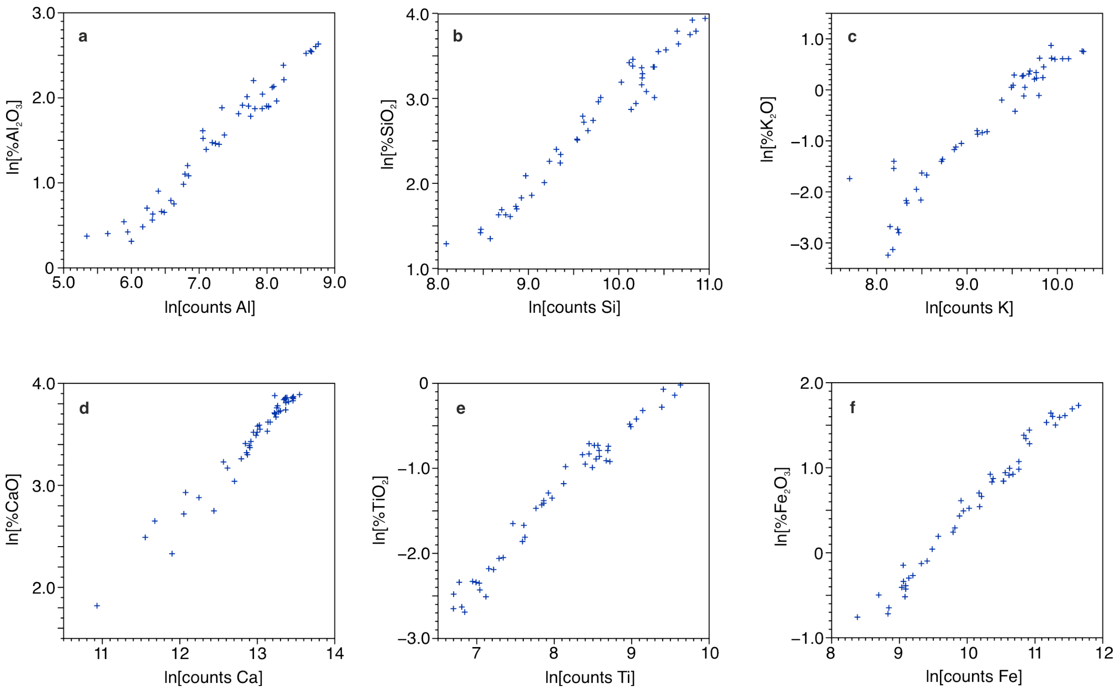
Supplementary Figure 4| Comparison of techniques for dust estimation at Site 659. Cross-plots of dust content as a percentage of the total sediment from Tiedemann et al. (1994)[2] plotted against: **a** % dust estimated by end-member unmixing (this study) and **b** ln[Ca/Fe] measured by XRF core scanning (this study). Measurements included in these plots were taken within 1 cm of each other. **c** Down core records of dust as a % of the total sediment by Tiedemann et al. 1994[2] (blue) and this study (red), from 0–5 Ma. All data have been converted onto the splice and age model developed in this study.



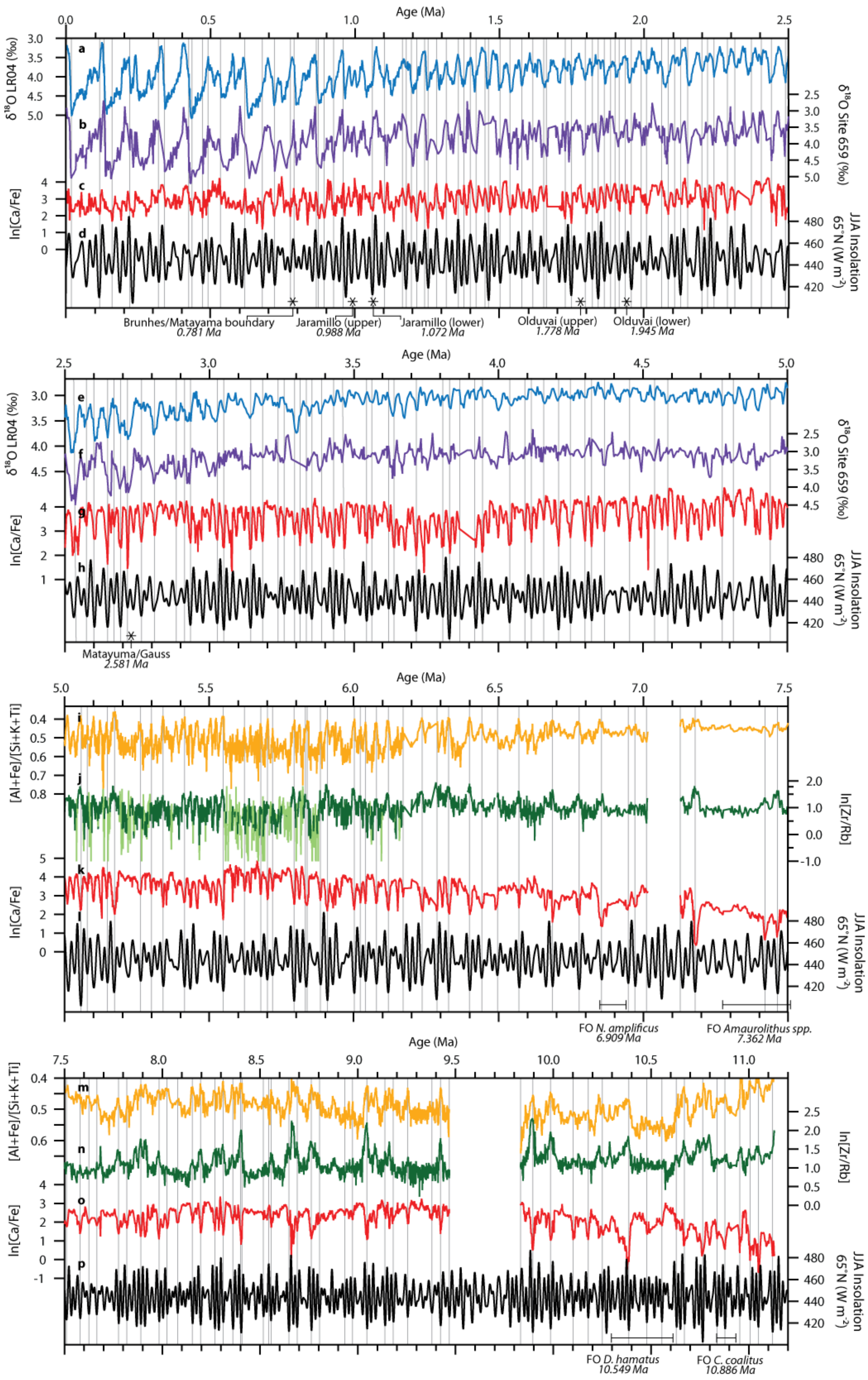
Supplementary Figure 5| Comparison of calibrated XRF ratios of Site 659 sediments spanning 0–11.2 Ma. **a** ln[Al/Si], **b** ln[Fe/K], **c** ln[Ti/Al], **d** [Al+Fe]/[Si+K+Ti], **e** ln[Ca/Fe]. Background image is a composited core photograph.



Supplementary Figure 6| Comparison of snapshots of calibrated XRF ratios of Site 659 sediment, with an example from each of zone I–III. Top: 0–1 Ma, middle 5–6 Ma, bottom 8–9 Ma. **a** ln[Al/Si], **b** ln[Fe/K], **c** ln[Ti/Al], **d** [Al+Fe]/[Si+K+Ti], **e** ln[Ca/Fe]. Background images are composited core photographs.



Supplementary Figure 7| Comparison of measurements of major element abundances in the calibration sample set between Avaatech XRF core scanner counts and oxide concentrations measured by discrete sample XRF analysis. **a** Al, **b** Si, **c** K, **d** Ca, **e** Ti and **f** Fe.



Supplementary Figure 8| Stratigraphic tie-points used in the construction of the Site 659 age model. **a,e** LR04 benthic foraminiferal oxygen isotope stack[61]. **b,f** Site 659 *C. wullerstorfi* oxygen isotope values[2] converted onto on our new composite depth scale and age model. **c,g,k,o** Calibrated ln[Ca/Fe] values of Site 659 sediments. **d,h,l,p** Mean boreal summer insolation (June-August) at 65°N given by the La2004 orbital solution[112]. **i,m** Calibrated

[Al+Fe]/[Si+K+Ti] ratios of Site 659 bulk sediment. **j,n** ln[Zr/Rb] values of Site 659 sediments (pale colour indicates low counts). Grey vertical lines mark the position of tie points used to generate the age model. The position of magnetostratigraphic boundaries as identified in the Leg 108 Scientific Results[108] are marked by stars, with listed ages from the Geologic Time Scale 2012 (GTS2012)[111], and biostratigraphic events (as identified in the Leg 108 Scientific Results[58] and J. Backman pers. comm.) are marked by bars, with ages updated following Raffi et al. (2006)[109]. Magnetostratigraphic and biostratigraphic dates were used to guide tuning.

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