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North Atlantic Color Cycles

1 **The transition on North America from the warm humid Pliocene to the**
2 **glaciated Quaternary traced by eolian dust deposition at a benchmark**
3 **North Atlantic Ocean drill site**

4

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21

22 **Abstract**

23 We present Plio-Pleistocene records of sediment color, %CaCO₃, foraminifer
24 fragmentation, benthic carbon isotopes ($\delta^{13}\text{C}$) and radiogenic isotopes (Sr, Nd, Pb) of
25 the terrigenous component from IODP Site U1313, a reoccupation of benchmark
26 subtropical North Atlantic Ocean DSDP Site 607. We show that (inter)glacial cycles
27 in sediment color and %CaCO₃ pre-date major northern hemisphere glaciation and are

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28 unambiguously and consistently correlated to benthic oxygen isotopes back to 3.3
29 million years ago (Ma) and intermittently so probably back to the Miocene/Pliocene
30 boundary. We show these lithological cycles to be driven by enhanced glacial fluxes
31 of terrigenous material (aeolian dust), not carbonate dissolution (the classic
32 interpretation). Our radiogenic isotope data indicate a North American source for this
33 dust (~3.3 to 2.4 Ma) in keeping with the interpreted source of terrestrial plant wax-
34 derived biomarkers deposited at Site U1313. Yet our data indicate a mid latitude
35 provenance regardless of (inter)glacial state, a finding that is inconsistent with the
36 biomarker-inferred importance of glaciogenic mechanisms of dust production and
37 transport. Moreover, we find that the relation between biomarker and the lithogenic
38 component of dust accumulation is distinctly non-linear. Both records show a jump in
39 glacial rates of accumulation from MIS G6 (2.72 Ma) onwards but the amplitude of
40 this signal is about 3 to 8 times greater for biomarkers than for dust and particularly
41 extreme during MIS 100 (2.52 Ma). We conclude that North America shifted abruptly
42 to a distinctly more arid and windy glacial regime from MIS G6, but major shifts in
43 glacial North American vegetation biomes and regional wind fields (exacerbated by
44 the growth of a large Laurentide ice sheet during MIS 100) likely explain
45 amplification of this signal in the biomarker records. Our findings are consistent with
46 wetter-than-modern reconstructions of North American continental climate under the
47 warm high CO₂ conditions of the Early Pliocene but contrast with most model
48 predictions for the response of the hydrological cycle to anthropogenic warming over
49 the coming 50 years (poleward expansion of the subtropical dry zones).

50

51 **Keywords:** Pliocene; Quaternary; eolian; dust; North America, North Atlantic;
52 Laurentide Ice Sheet.

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53

54 **1. Introduction**

55 Deep-sea sediments in the climatically sensitive North Atlantic region are composed
56 of two main constituents: biogenic carbonate (CaCO_3) produced in the overlying
57 water column and allochthonous detrital material, with volcanic ash important only
58 locally. It has long been recognised that striking rhythmic changes in the abundance of
59 these constituents and therefore sediment color and $\% \text{CaCO}_3$ (Figure 1) provide both
60 a high fidelity means of stratigraphic correlation and an expression of pronounced
61 climate variability, especially in sediments deposited during times of significant
62 northern hemisphere glaciation (NHG) (e.g. Ericson et al., 1961; Ruddiman and
63 Glover, 1972).

64 Shackleton et al. (1984) drew attention to the remarkable correspondence
65 between high amplitude changes in both benthic $\delta^{18}\text{O}$ and $\% \text{CaCO}_3$ back to earliest
66 Pleistocene Marine Isotope Stage (MIS) 100 (2.52 Ma) at Deep Sea Drilling Project
67 (DSDP) Site 552, where sediment deposition is dominated by pelagic rain from
68 above. Prior to 2.52 Ma, variance in benthic $\delta^{18}\text{O}$ is unaccompanied by large
69 amplitude change in $\% \text{CaCO}_3$ at Site 552 (Figure 1A). Originally, initiation of high-
70 amplitude variance in color and $\% \text{CaCO}_3$ at this site was attributed by Shackleton et
71 al. (1984) to onset of major NHG with $\% \text{CaCO}_3$ controlled by variations in the flux of
72 non-carbonate material transported by ice rafting. We now know that the exact timing
73 of the large decrease in $\% \text{CaCO}_3$ in sediments deposited at Site 552 is obscured
74 because MIS G6 through 103 (2.72–2.58 Ma) fall in a core break (Raymo et al.,
75 1989). As it happens, however, extensive work elsewhere shows MIS 100 to be the
76 oldest glacial during which ice sheets were large enough (Ruddiman et al. 1987;
77 Maslin et al. 1998; Jansen and Sjöholm, 1991; Jansen et al. 2000; Kleiven et al. 2002;

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78 Bailey et al., 2013) and high latitude surface ocean temperatures cool enough
79 (Lawrence et al. 2009; 2010; Naafs et al. 2010) to initiate ice-rafting on a basin-wide
80 scale across the open North Atlantic Ocean.

81 In Figure 1A we show %CaCO₃ records from two further classic North
82 Atlantic drill sites, DSDP 607 and 609 located on the southern fringe and at the centre
83 of the last glacial IRD belt, respectively (Figure 2). Originally, %CaCO₃ variability at
84 these two sites prior to ~2.5 Ma was attributed to sea floor CaCO₃ dissolution, a
85 consequence of their greater water depth (Sites 609, ~3.9 km & 607, ~3.5 km, vs. 552
86 ~2.3 km, Figure 1A) and the influence of corrosive poorly ventilated southern-
87 sourced bottom waters (Ruddiman et al., 1987; Ruddiman and Raymo, 1988). Yet,
88 comparison of the %CaCO₃ plots compiled by Ruddiman et al. (1987) to records from
89 shallower more recently drilled sites (Figure 1A vs. 1B) reveals that the timing of the
90 initiation of marked lithological cycles in North Atlantic Ocean sediments is not a
91 simple function of water depth indicating the influence of some factor other than
92 CaCO₃ dissolution.

93 In principle, three mechanisms have the potential to deliver terrigenous
94 sediments to Site U1313. But negligible Pliocene rates of accumulation of sand-sized
95 IRD and volcanic grains at Integrated Ocean Drilling Program (IODP) Site U1313
96 (Bolton et al. 2010), the reoccupation of DSDP Site 607, confirms that the
97 contemporaneous variability seen in %CaCO₃ (Figure 1) is not a function of melting
98 icebergs over this classic site. Two alternative potential explanations must therefore be
99 considered: (1) Transport beyond the contemporary iceberg front by ocean currents of
100 fine-grained material delivered by ice-rafting to the Nordic Seas by the Greenland Ice
101 Sheet (Winkler, 1999; Jansen et al., 2000; Andrews 2000). (2) Transport of
102 continentally derived eolian dust from North Africa or from North America as inferred

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103 based on biomarker records (Naafs et al., 2012).

104 To better understand the control(s) on, and climatic significance of, %CaCO₃
105 variability of North Atlantic Ocean sediments deposited during the intensification of
106 northern hemisphere glaciation (iNHG) we present new orbital-resolution records of
107 carbonate dissolution, benthic $\delta^{13}\text{C}$, coarse lithic abundance and sediment %CaCO₃
108 for IODP Site U1313, and radiogenic isotopes datasets that track the provenance of
109 terrigenous inputs to this site. We show that lithological cycles in North Atlantic
110 sediments of Pliocene age are driven by enhanced glacial fluxes of terrigenous
111 material, not carbonate dissolution. Our provenance work indicates that the
112 terrigenous component at the site is dominated by eolian dust sourced from the mid
113 latitudes of North America – a result consistent with published interpretations of the
114 record from Site U1313 of biomarkers derived from higher plant leaf waxes (Naafs et
115 al., 2012). A sharp increase in the biomarker proxy for dust inputs to our study site
116 during MIS G6, 2.72 Ma, is interpreted to reflect the importance of glacial grinding
117 by a large North American ice sheet complex in amplifying dust inputs to the North
118 Atlantic Ocean during glacials from this time (Naafs et al., 2012). Comparison among
119 data sets, however, indicates strong non-linearity in coupling between the dust and
120 biomarker records indicating that a reappraisal is merited of the sequence of climatic
121 events that they record and the mechanisms involved.

122

123 **2. Materials & Methods**

124 *2.1. IODP Site U1313*

125 IODP Site U1313 is located at the base of the upper western flank of the Mid Atlantic
126 Ridge at a water depth of 3426 m, ~240 nautical miles northwest of the Azores
127 archipelago (41°N, 32.5°W), on the extreme southerly limit of the last glacial “IRD

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128 belt” (Ruddiman, 1977), a southwest-northeast trending band of maximum iceberg
129 melting and hence IRD deposition between ~40°N and 55°N in the Atlantic Ocean
130 (Figure 2). Site U1313 was drilled during IODP Expedition 306 and constitutes a
131 reoccupation of DSDP Site 607, a benchmark mid-depth site monitoring North
132 Atlantic Deep Water throughout the Plio-Pleistocene (Ruddiman et al., 1987; Raymo
133 et al., 1989; Ruddiman et al., 1989; Raymo et al., 1992; Raymo et al., 2004). Site
134 U1313 offers distinct advantages over its Site 607 precursor because it benefits from
135 recovery by modern coring methods and from application of a full suite of physical
136 property data collection and stratigraphic correlation techniques (Channel et al.,
137 2010).

138

139 *2.2. Stable isotope analysis, foraminifera fragmentation and chronology*

140 Samples from IODP Site U1313 were obtained at 10 cm resolution from 114.12 to
141 155.28 meters composite depth (mcd) and washed over a 63 μm sieve. The ratio
142 between fragments and whole foraminifera was established for the >150 μm fraction
143 where more than 300 whole foraminifera were present following Ivanova et al.
144 (2003). We focus our discussion on the interval 3.33 Ma (MIS MG1) to 2.41 Ma (MIS
145 95) covered by a published oxygen isotope stratigraphy (Bolton et al., 2010),
146 measured on the benthic foraminifera *Cibicidoides wuellerstorfi* (>212 μm). For
147 discussions of the younger Pleistocene portion of the Site U1313 record, we utilize the
148 age model of Naafs et al. (2012) based on benthic $\delta^{18}\text{O}$ datasets spanning three time
149 windows of the past 1 Myr (Stein et al., 2009; Ferretti et al., 2010; Naafs et al., 2012)
150 and shipboard correlation (Expedition 306 Scientists, 2006) of sediment physical
151 properties (L^* , lightness) to the LR04 stack (Lisiecki and Raymo, 2005). We present a
152 new benthic $\delta^{13}\text{C}$ record from the samples analysed by Bolton et al. (2010) with an

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153 external analytical precision, based on replicate analysis of an in-house standard
154 calibrated to NBS-19, of $\pm 0.031\%$ (1σ).

155

156 *2.3 Coarse lithic counts*

157 The only high-resolution record of IRD deposition at Site U1313 for iNHG spans MIS
158 102-96 (Bolton et al., 2010). To improve our understanding of the history of IRD
159 deposition at our study site during MIS M2 (3.3 Ma) and between MIS G6 and MIS
160 102 (2.72–2.56 Ma), new coarse lithic counts were performed on the $>150\mu\text{m}$ size
161 fraction between, respectively, 152.98–154.98 mcd and 120.74–129.86 mcd. The
162 abundance of coarse lithics in Site U1313 sediments are very low throughout our
163 study interval and are extremely low in sediments deposited prior to MIS 100 (2.52
164 Ma). To generate a statistically significant record of sand IRD abundance in Site
165 U1313 sediments (expressed as IRD per gram of dry sediment) we therefore counted
166 all coarse IRD ($>150\mu\text{m}$) in each sample studied.

167

168 *2.4. Sediment color*

169 There is a long history of attempts to develop rapid-throughput proxy methods to
170 estimate sediment $\%\text{CaCO}_3$ from the spectral properties of sediments (e.g. Chester
171 and Elderfield, 1966). Use of optical lightness as an analytical tool in sediments
172 recovered by DSDP and IODP has its roots in late Quaternary studies (Balsam, 1981),
173 was pioneered by grey-scale analysis of photographs (e.g. Herbert and D'Hondt,
174 1990; Busch, 1991) and is now determined routinely from sediment color in
175 sediments of appropriate lithology. Sediment color can be defined using three
176 variables, a^* (red-green), b^* (blue-yellow) and L^* (lightness), that lie along mutually
177 perpendicular axes in color space. We obtained shipboard color reflectance data at 2

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178 cm resolution for Site U1313 from the IODP database website
179 (<http://iodp.tamu.edu/database/index.html>), generated using a modern version of the
180 split core automatic track reflectance spectrometer first trialled to remarkable effect
181 during Ocean Drilling Program (ODP) Leg 138 (Mix et al., 1992; Mix et al., 1995;
182 Ortiz et al., 1999). Here we employ records of L^* to represent sediment color.

183 A comparison of L^* data for Site U1313 and discrete %CaCO₃ measurements
184 generated post-cruise on sediments deposited during MIS 16–9 (640–320 ka; Stein et
185 al., 2009) illustrates that large variations in L^* for Site U1313 sediments deposited
186 over late Pleistocene glacial-interglacial cycles correspond to pronounced variations
187 (of ~30 %) in sediment %CaCO₃ (Figure 3). To improve our understanding of the
188 relationship between sediment color and %CaCO₃ for the Pliocene portion of the Site
189 U1313 record (for which L^* values are typically higher and amplitude change muted
190 relative to those documented for the Pleistocene) we generated 193 new %CaCO₃
191 estimates on small (~0.5 cc), discrete samples using a standard (LECO) combustion
192 technique following Stein et al. (2009).

193 To generate a high-resolution record of %CaCO₃ of Site U1313 sediments for
194 the past 3.3 Ma, we perform a least-squares linear regression between our new
195 discrete %CaCO₃ data (Figure 3C; n = 193), supplemented by the previously
196 published %CaCO₃ data (Figure 3B; n = 151 (Stein et al., 2009)), and 10 cm (5 point)
197 running average of the L^* data series (Figure 3D). The excellent linear correlation
198 ($r^2=+0.88$, $p<0.001$; Figure 3D) between these two variables indicates that our orbital-
199 resolution L^* -based estimates of %CaCO₃ are not strongly influenced by potential
200 complicating factors (e.g. changing composition of the non-CaCO₃ fraction (Balsam
201 et al., 1999)). This calibration is applicable to the task of generating a record of
202 %CaCO₃ for Site U1313 sediments of Pliocene through late Pleistocene age, but the

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203 resultant %CaCO₃ record can only be used to estimate eolian dust fluxes prior to the
204 late Pleistocene interval because of the error propagation associated with notable
205 delivery of IRD during late Pleistocene ice-rafting events, most notably the extreme
206 Heinrich events (see Section 3.3 (Stein et al., 2009; Naafs et al., 2013)). Fortunately,
207 our focus is on the origin and temporal evolution of terrigenous MARs at our study
208 site during the Pliocene where the linear fit is excellent and our new IRD record
209 demonstrates that the terrigenous sediment component contains negligible (i.e.
210 interglacial-like) ice-rafted sand-sized grains.

211

212 *2.5. Radiogenic isotope data*

213 The radiogenic isotope (Nd, Pb, Sr) composition of Atlantic Ocean sediment is well
214 established as a tracer of both eolian sediment (e.g. Grousset et al., 1998; Abouchami
215 and Zabel, 2003; Grousset and Biscaye, 2005) and ice rafted material (e.g. Revel et
216 al., 1996; Grousset et al., 2001; Fagel et al., 2002; Fagel et al., 2004; Fagel and
217 Matielli, 2011; Colville et al., 2011). These applications rely on regional differences in
218 circum-North Atlantic Ocean geology as a function of age and tectonic (metamorphic)
219 history.

220 To understand better the origin of the terrigenous component of Site U1313
221 Pliocene sediments, we have measured the Pb, Nd and Sr isotopic composition of
222 carbonate-free bulk terrigenous samples selected from peak glacials and interglacials
223 associated with the interval of iNHG (3.5–2.5 Ma; Mudelsee and Raymo (2005)).
224 Sample processing closely followed Gutjahr et al. (2007). Approximately 0.5 g of
225 crushed and homogenised bulk sediment was decarbonated using a Na acetate buffer,
226 and absorbed metals were removed with a 1M MgCl₂ solution. Authigenic coatings
227 were then removed using a 0.05 M hydroxylamine hydrochloride – 15 % acetic acid

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228 – 0.03 M Na–EDTA solution buffered to pH 4 with analytical grade NaOH in two
229 steps totalling 27 hours on a shaker table. Following removal of organic matter using
230 hydrogen peroxide and aqua regia, samples were pressure-dissolved in a HF-HNO₃
231 mixture.

232 Pure samples of Pb, Nd and Sr were extracted using standard procedures. The
233 Nd-isotope (¹⁴³Nd/¹⁴⁴Nd) and Pb-isotope ratios (²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb and
234 ²⁰⁸Pb/²⁰⁴Pb) of our processed samples were measured at the University of
235 Southampton using a multi-collector inductively coupled plasma mass spectrometer
236 (MC-ICP-MS, Thermo Scientific Neptune). Neodymium isotopic compositions were
237 obtained using the method of Vance and Thirlwall (2002) through adjustment to a
238 ¹⁴⁶Nd/¹⁴⁴Nd value of 0.7219. Mass-bias corrected ratios were normalized to the given
239 ¹⁴³Nd/¹⁴⁴Nd value (0.512115) of the standard JNdi-1 (Tanaka et al., 2000). Mass bias
240 corrected Pb isotopic compositions were measured following a standard-sample
241 bracketing approach normalizing Pb isotopic compositions of NBS981 to the values
242 of Baker et al. (2004). The Strontium isotope composition (⁸⁷Sr/⁸⁶Sr) of these samples
243 was also measured at the University of Southampton using a thermal ionisation mass
244 spectrometer (ThermoFisher TRITON Plus). Total procedural blanks averaged 174pg,
245 106pg and 195pg for Nd, Sr and Pb, respectively. External precisions are calculated
246 (at 2 standard deviations) as the reproducibility of the following standards: JNdi-1
247 (Nd), NBS 987 (Sr) and NBS 982 (Pb). Precision is 0.000007 (<0.15 εNd), 0.000015,
248 0.047, 0.022 and 0.062 for ¹⁴³Nd/¹⁴⁴Nd, ⁸⁷Sr/⁸⁶Sr, ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb and
249 ²⁰⁸Pb/²⁰⁴Pb respectively. For convenience Nd isotope ratios are reported in epsilon
250 notation as:

$$\epsilon_{Nd} = \left[\frac{{}^{143}\text{Nd}/{}^{144}\text{Nd}_{\text{sample}}}{{}^{143}\text{Nd}/{}^{144}\text{Nd}_{\text{CHUR}}} - 1 \right] \times 10^4$$

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253 where $^{143}\text{Nd}/^{144}\text{Nd}_{\text{CHUR}}$ reflects the Chondrite Uniform Reservoir value of 0.512638

254 (Jacobsen and Wasserburg, 1980).

255 We assessed the provenance of terrigenous sediments deposited at Site U1313

256 by comparing their Pb, Sr and Nd isotopic compositions to equivalent radiogenic

257 isotopic compositions of potential source regions, which are based on our compilation

258 of discrete measurements made on circum-North Atlantic Ocean bedrock, terrestrial

259 loess outcrop, atmospheric aerosols, and continental ice and dust source-proximal

260 (core top and down-core) marine sediments and river samples (Figure 2a and 4 and

261 Supplementary Information). Potential source areas for IRD deposited in the North

262 Atlantic Ocean fall into three groups marked by a range of radiogenic isotope

263 compositions (Figure 2a and 4 (c.f. Thierens et al., 2012)). The old, primarily

264 Precambrian terranes of Greenland and North Eastern Canada (including the Labrador

265 Sea, Hudson Strait and Baffin Bay) comprise the “Canadian Province” (Dawes et al.,

266 2009). Paleocene to recent volcanic rocks found in Eastern Greenland, Iceland and the

267 Faeroe Islands comprise the “Volcanic Province”, local Azores volcanism may also

268 contribute material of this composition. Together, areas with their corresponding

269 compositions represent the high-latitude regions that constitute the most likely sites of

270 early ice sheet growth (e.g. Winkler et al., 1999; DeConto et al., 2008). Lower-latitude

271 ice rafting from Britain, Scandinavia or North America (the Appalachian terrane and

272 Grenville Province in the region of the Gulf of Saint Lawrence) were important

273 sources of ice-rafted material to the North Atlantic Ocean during the last glacial

274 maximum (Watkins et al., 2007). Owing to their similarities in Pb and Nd-Sr isotope

275 spaces, we group these three distinct geographic regions into a third province,

276 intermediate in age to the two high-latitude provinces. Eolian material sourced from

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277 the Sahara and North America has a similar geologic age and isotopic composition to
278 the Fenoscandinavian tectonic terranes and the Gulf of St Lawrence region of North
279 America, but is unequivocally distinct from high-latitude Volcanic Province material
280 and Precambrian and Proterozoic Canadian and Greenland terranes in Nd-Sr space.

281

282 **3. Results and Discussion**

283 *3.1. Stable isotope stratigraphy and sediment color*

284 The record of benthic $\delta^{13}\text{C}$ at Site U1313 shows only modest glacial-interglacial
285 variability with the exception of prominent excursions to low values during the large
286 benthic $\delta^{18}\text{O}$ glacials MIS 100, 98 and 96 (Figure 5). This result is consistent with the
287 record from predecessor Site 607 (Raymo et al., 1989), but the prominent
288 (inter)glacial $\delta^{13}\text{C}$ signal established in MIS 100 is more pronounced in our record.
289 Our record also resolves with higher fidelity earlier key glacials and illustrates, for
290 example, that MIS M2 (~3.3 Ma), the first prominent excursion in benthic $\delta^{18}\text{O}$ to
291 interrupt early Pliocene warmth, is not associated with a prominent benthic $\delta^{13}\text{C}$
292 excursion indicative of corrosive southern sourced waters.

293 Our L^* -derived record of sediment $\% \text{CaCO}_3$ at Site U1313 is shown in
294 Figures 1A and 5 and reveals the expected North Atlantic pattern (lighter, CaCO_3 -rich
295 sediments during interglacials and darker more terrigenous-rich intervals during
296 glacials), but the fidelity of the signal and its unambiguous correlation to our benthic
297 $\delta^{18}\text{O}$ series are remarkable back to 3.3 Ma (the base of our isotope record – Figure
298 5G). This relationship was postulated for Site U1313 based on shipboard correlation
299 of L^* to the LR04 stack (Expedition 306 Scientists, 2006). Here we confirm, using
300 our co-registered signal ($\% \text{CaCO}_3$ and benthic $\delta^{18}\text{O}$ determined from the same
301 sediments) that variations in L^* at Site U1313 track changes in benthic $\delta^{18}\text{O}$ at this

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302 site across iNHG from 3.33 to 2.4 Ma. This result demonstrates that the onset of clear
303 glacial-interglacial lithological cycles at this site took place at least 800 kyr earlier
304 than the onset both of basin-wide ice rafting at MIS 100, 2.52 Ma, and of pronounced
305 glacial-interglacial variability in benthic $\delta^{13}\text{C}$ at our study site (Figure 5B).

306

307 *3.2. Abundance of the carbonate sedimentary component at Site U1313*

308 Pliocene sediments at Site U1313 are characterized by small variations in color and
309 %CaCO₃ relative to the higher amplitude changes that characterize the late
310 Pleistocene (Figure 1). High amplitude changes in Pleistocene %CaCO₃ (and color)
311 from the North Atlantic Ocean are often interpreted to reflect primarily changes in
312 carbonate production and dilution by other sediment components (e.g. Lototskaya et
313 al., 1998; Helmke and Bauch, 2001), while the lower amplitude %CaCO₃ variations
314 observed during the Pliocene at DSDP Sites 607 and 609 have been classically
315 attributed to dissolution on glacial-interglacial timescales (Ruddiman et al., 1987).
316 Our analysis, however, calls this classic interpretation into question. Calcareous
317 microfossils are extremely well preserved in Pliocene sediments from Site U1313
318 with foraminifera fragment counts typically well within the zero ΔCO_3 range of Le
319 and Shackleton (1992) (Figure 5A). The relationship between the fraction of CaCO₃
320 dissolved and that remaining is highly non-linear such that, when the CaCO₃ fraction
321 is large, substantial CaCO₃ must be dissolved to achieve small percentage variations
322 (Berger, 1971). For example, to generate a change in carbonate content of the order
323 observed at Site U1313/607 between about 3.3 and 2.8 Ma (~95% to 85%, Figure
324 5G), about 60% of the initial CaCO₃ must be dissolved. Such substantial dissolution
325 of CaCO₃ at Site U1313 is not consistent with the extremely well preserved
326 calcareous microfossils observed in these sediments. Thus, in contrast to the classic

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327 interpretation, CaCO₃ dissolution does not control carbonate content at Site
328 U1313/607 prior to MIS 100 and cannot be used to assess changes in North Atlantic
329 deep-water carbonate chemistry through time. Instead, the dominant controls must be
330 calcite production and/or terrigenous dilution (Ruddiman and McIntyre, 1976;
331 Ruddiman et al., 1987; Lototskaya et al., 1998; Helmke and Bauch, 2001).

332 A recently published record of alkenone accumulation from Site U1313 (Naafs
333 et al., 2010) reveals the onset of high amplitude glacial-interglacial changes in
334 alkenone accumulation, and therefore total export productivity (Bolton et al., 2010b;
335 Bolton et al., 2011), from ~2.72 Ma (MIS G6). The orbital signal in the alkenone data,
336 however, is of the wrong sign for carbonate productivity to control sediment color and
337 CaCO₃ burial (alkenone accumulation peaks during glacials whereas %CaCO₃ and
338 color, L*, peak during interglacials; Figure 5C). Furthermore, our records demonstrate
339 that terrigenous accumulation peaks during glacials throughout our study interval and
340 not just from ~2.72 Ma onwards (Figure 5E). We conclude that the glacial-interglacial
341 signal in Pliocene sediment color and %CaCO₃ at Site U1313 is driven by addition of
342 terrigenous material. Next we assess the potential mechanisms by which this
343 terrigenous material might have been transported to our study site.

344

345 *3.3. Radiogenic isotopes and sediment provenance*

346 The Sr, Nd and Pb isotope composition of the bulk sediment terrigenous fraction
347 deposited at Site U1313 during peak interglacial and glacial conditions during iNHG
348 are shown in Figure 6. The Sr and Pb isotope composition of the samples analysed
349 display a relatively small range of variability, with ⁸⁷Sr/⁸⁶Sr ranging from 0.71664 to
350 0.72561 and ²⁰⁶Pb/²⁰⁴Pb ranging from 18.20 to 18.97 (Fig 7A and C, respectively).
351 Variation in εNd is more pronounced (ranging between -9.85 to -17.67), although

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352 most values fall between -13.9 and -16 (Figure 7B). Two samples (corresponding to
353 interglacials MIS G1 and 101) are indicative of volcanic material transported from
354 East Greenland or Iceland, or from the Azores volcanic islands. Remarkably, aside
355 from these two volcanically influenced exceptions, Nd, Sr and Pb isotope ratios show
356 no systematic difference between samples selected from peak glacial and peak
357 interglacial climate states (across a range in benthic $\delta^{18}\text{O} > 1.5\text{‰}$, Fig. 7D).

358 Based on the continuous presence of terrestrial leaf waxes in the Pliocene
359 sediments at our study site (a tracer of eolian dust in marine sediments at Site U1313;
360 Figure 5D), we know that at least some portion of the terrigenous fraction at Site
361 U1313 is composed of eolian dust. This biomarker record exhibits fluxes akin to those
362 observed for wind-blown leaf-waxes deposited in the Southern Ocean over the past ~4
363 Ma - where eolian dust is known to dominate the make-up of terrigenous sediments at
364 ODP Site 1090 (Martínez-García et al., 2011). Comparison of the Nd, Sr and Pb
365 isotope composition of Site U1313 terrigenous sediments to those of potential source
366 regions points to a definitive (non-volcanic) mid-latitude origin (Figures 6 and 7 and
367 Supplementary Information). This observation is paleoclimatically powerful because
368 it demonstrates that, prior to MIS G6 (>2.72 Ma), in a world characterized by only
369 incipient high latitude NHG (e.g. Bintanja and van de Wal, 2008), eolian dust supply
370 from the Sahara or North America is the only credible source capable of producing
371 orbital-scale cyclical variations in terrigenous inputs to our study site. Our data are
372 incompatible with a contribution from Greenland or Northern Canada, which are
373 widely inferred to have been the nucleation points of the earliest northern hemisphere
374 ice sheets (Winkler, 1999; Jansen et al., 2000; DeConto et al., 2008). In fact, where
375 present, sand-sized IRD and volcanic grains occur in only trace numbers (typically 0
376 to <0.1 grains/g) in sediments older than MIS G4 at Site U1313. This finding,

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377 together with the lack of a glacial-interglacial signal in our geochemical data, makes a
378 high-latitude or even an improbable mid-latitude glacial origin for the bulk
379 terrigenous fraction at Site U1313 untenable for sediments deposited prior to 2.7 Ma.
380 These findings indicate that the bulk terrigenous sediment component deposited at
381 Site U1313 between 3.3 Ma and 2.7 Ma is dominated by eolian dust.

382 We might expect a direct contribution from ice rafting to the terrigenous
383 sediment component at Site U1313 during glacials between ~ 2.72 Ma and 2.4 Ma
384 associated with the onset of significant NHG (Kleiven et al., 2002). As in the case of
385 the older part of our record, however, our grain counts reveal, accumulation rates of
386 sand-sized IRD in glacial sediments from MIS G4 onwards to be negligible (Figure
387 5). This result is in keeping with the location of Site U1313, situated far south
388 ($\sim 41^\circ\text{N}$) of the late Pliocene IRD belt (centred on $\sim 53^\circ\text{N}$; Bailey et al., 2013) and on
389 only the southernmost fringe of the IRD belt even during the Last Glacial (Figure 1).
390 This finding, together with, the consistent mid-latitude geochemical provenance
391 indicated for terrigenous sediments deposited at Site U1313 leads us to conclude that,
392 throughout our Pliocene record, the contribution made by IRD deposition shed from
393 icebergs over site to the average radiogenic isotope composition of terrigenous
394 sediments is insignificant in comparison to the major player, eolian dust. Similarly,
395 despite evidence for abundant deposition of IRD in the higher latitude northeast North
396 Atlantic Ocean sourced from high-latitude Archaean and Proterozoic-aged terranes
397 during glacials since ~ 2.72 Ma (Bailey et al., 2013), the consistent mid-latitude
398 geochemical provenance that we report here for the terrigenous fraction at Site U1313
399 makes it extremely unlikely that transport of fine-grained IRD beyond the
400 contemporary iceberg front by ocean currents could be responsible for terrigenous
401 deposition at our study site from MIS G6 onwards.

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402 Similarities of the Nd and Sr isotopic composition of North American and
403 Saharan eolian dust means that we must consider additional lines of evidence to pin
404 down the source of the dust at Site U1313. Today, while most of the African-derived
405 dust is driven westwards over the tropical North Atlantic by the Trade Winds, some
406 dust is also transported northwards towards the North Atlantic, the Mediterranean and
407 as far north as Northern Europe (Bergametti et al., 1989; Moulin et al., 1997; Kuss
408 and Kremling, 1999; Kellog and Griffin, 2006). Three lines of evidence, however,
409 support the notion that eolian dust deposition at our study site during iNHG is
410 dominated by North American sources. First, spectral analysis of our record of
411 terrigenous accumulation at Site U1313 reveals a dominant obliquity beat (with no
412 strong precession signal) throughout our study interval that is in contrast to the pattern
413 of variability in records of Saharan dust deposition (Fig. 8). Saharan dust deposition
414 reveals the influence of obliquity from ~2.7 Ma, but precessional variability is
415 important for at least the last 5 Ma (Tiedemann et al., 1994; DeMenocal, 2004)).
416 Second, both modern wind trajectories (Figure 2B) and those modelled for both the
417 last glacial maximum and Pliocene (Haywood et al., 2000; Hewitt et al., 2003;
418 Pausata et al., 2011) indicate that Site U1313 is strongly influenced by intense
419 westerly winds originating from the major present-day North American dust source
420 region, the American Southwest (including all land between 125°W and 95°W and
421 25°N and 40°N) that incorporates the southwestern United States and parts of
422 northern Mexico (Seager et al., 2007). Third, organic biomarker- and clay
423 mineralogy-based provenance studies, respectively, independently link Plio-
424 Pleistocene eolian derived terrestrial high plant waxes at Site U1313 (Naafs et al.,
425 2012) and Holocene eolian-derived material across the central North Atlantic
426 (Grousset and Chesselet, 1986) to North American sources.

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427

428 *3.4. Records and mechanisms of North American eolian dust flux during Pliocene*

429 *iNHG*

430 Dust is both a signal and an agent of climate change (Martin et al., 1990; Kohfield and
431 Harrison, 2001; Mahowald et al., 2005; Winckler et al., 2008; Ganopolski et al., 2010;
432 Sun et al., 2010; McGee et al., 2010). To date, however, nearly all we know of the
433 history of eolian dust export from North America during Pliocene iNHG comes from
434 a single proxy biomarker record of terrestrial higher plant leaf wax (organic n-alkane)
435 deposition at Site U1313 (Naafs et al., 2012). That benchmark high-resolution record
436 shows that eolian-derived n-alkane and alkan-1-ols inputs to the North Atlantic Ocean
437 jumped to higher glacial values from ~2.7 Ma (MIS G6, Figure 5). Yet, these
438 biomarkers represent only a minor and often highly variable component of eolian dust
439 (Huang et al., 2000; Conte and Weber, 2003) so it is important to compare the proxy
440 biomarker record for dust deposition at Site U1313 with our record of variations in the
441 deposition of the terrigenous sediment component at this site.

442 Our record reveals that, while dust fluxes to our study site prior to the onset of
443 significant NHG at ~2.72 Ma are lower than those associated with Quaternary
444 glaciations, they are still high (up to 0.9 g/cm²/ka) and unambiguously mimic global
445 climate (as recorded by benthic $\delta^{18}\text{O}$) back to at least 3.3 Ma (Figure 5E). The two
446 published biomarker records reveal that, from ~2.7 Ma onwards, glacial accumulation
447 of the organic fraction of North American dust increases significantly during glacials
448 at Site U1313 (Figure 5D). We are careful not to interpret our terrigenous record as a
449 pure signal of lithogenic dust deposition from this time because our records show
450 evidence for a contribution from ice rafting during glacials from MIS G4 onwards
451 (albeit extremely small, see Section 3.3). But our record of the bulk terrigenous flux

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452 to Site U1313 places an upper limit on the potential magnitude of increase in eolian
453 dust flux to our study site that is possible during MIS G6 relative to background
454 interglacial values prior to this time: it can not be greater than a factor of about two
455 (Figure 5E). In fact, the peak fluxes that we record for the bulk terrigenous fraction
456 around 2.7 Ma ($\sim 2 \text{ g/cm}^2/\text{ka}$, Figure 5E) are similar to the lower end of the estimated
457 range of late Pleistocene glacial dust flux to the North Atlantic Ocean ($2\text{-}5 \text{ g/cm}^2/\text{ka}$;
458 Maher and Denis, 2001).

459 The jumps in glacial accumulation in both the biomarker records and our
460 terrigenous record from MIS G6 (2.72 Ma) onwards (Figures 5D and 5E) strongly
461 suggest that the North American continent shifted abruptly into a distinctly more
462 Pleistocene-like cold stage regime (cold, arid, and windy) from MIS G6. One
463 potential mechanism for the sudden jump in dust inputs to our study site from 2.72
464 Ma is the development of large ice-sheets on North America from MIS G6 onwards as
465 inferred by Naafs et al. (2012). Large ice sheets advancing over regolith-rich Pliocene
466 terrains (Clark and Pollard, 1998) provide an attractive mechanism for delivering fine-
467 grained sediments to mid-latitude outwash plains for eolian entrainment (Ganopolski
468 et al., 2010), but three lines of evidence call this interpretation into question. (1)
469 Typically, the biomarker component of atmospheric dust becomes wind-entrained
470 through ablation from living vegetation assisted by sand blasting (eolian abrasion)
471 rather than by the deflation of soils and glacial outwash plains in dust source regions
472 (Huang et al., 2000; Conte and Weber, 2002; Schefuß et al., 2003). (2) Recent work
473 on the provenance of North Atlantic IRD (Bailey et al. 2013) and on Arctic climate
474 (Bringham-Grette, 2013) during iNHG indicates that major ice sheets (i.e. extending
475 into the mid latitudes) are unlikely to have been sustained in North America as early
476 as during MIS G6. In fact, our data show that dust inputs to Site U1313 were likely

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477 substantial during cold stages well before MIS G6 with a particularly prominent peak
478 in MIS M2 (Figure 5E), long before the existence of a large Laurentide ice sheet is
479 tenable (De Schepper et al., 2013; Bringham-Grette et al., 2013). (3) Our radiogenic
480 isotope data show a consistently mid-latitude provenance of the bulk terrigenous
481 fraction at Site U1313 from 3.3 through 2.4 Ma regardless of glacial-interglacial state,
482 thereby ruling out a significant high latitude contribution, even during MIS 100.
483 These observations suggest that non-glaciogenic processes of Pliocene dust
484 production, akin to those important during the last glacial maximum (e.g. increased
485 wind intensity, enhanced aridity and reduced vegetation (Rea et al., 1994; Aleinikoff
486 et al., 1999; Mason 2001; Werner et al., 2002; Bettis et al., 2003; Winkler et al., 2002;
487 Prospero et al., 2002; Bussaca et al., 2003 Mahowald et al., 2006; Aleinikoff et al.,
488 2008; McGee et al., 2010)), are more important than suggested previously.

489

490 *3.5. Non-linearity in the relation between biomarkers and terrigenous eolian dust*
491 *deposition in the North Atlantic Ocean during Pliocene intensification of northern*
492 *hemisphere glaciation.*

493 In Figure 9 we present cross plots of our record of terrigenous mass accumulation at
494 Site U1313 and the published biomarker records of Naafs et al. (2012) for our study
495 interval (3.33-2.41 Ma). These cross plots reveal a close association between
496 biomarker and terrigenous sediment accumulation at our study site but, in contrast to
497 what is seen in other paleo-dust proxy records (Winckler et al., 2008) including other
498 applications of the n-alkane technique (e.g. at South Atlantic Site 1090; Figure 9C;
499 Martinez-Garcia et al., 2011), the relationships observed between the biomarkers and
500 terrigenous fraction in Site U1313 are distinctly non-linear (e.g. Figure 9A vs. 9B).
501 While stratigraphic comparison of these three records shows that they all display

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502 jumps in glacial accumulation from MIS G6 (2.72 Ma) onwards (Section 3.4), the
503 amplitude of this signal is about 3 to 8 times greater for biomarkers than for
504 terrigenous inputs (Figure 10). The amplified jump in the biomarker records relative
505 to the jump in the terrigenous record is particularly extreme during MIS 100 (2.52
506 Ma), especially in the record of n-alkan-1-ol accumulation (Figure 10). This
507 observation underscores an important point: Non-linearity in the relation between the
508 biomarkers and the lithogenic record cannot be explained by invoking the input of
509 terrigenous material through additional mechanisms (ice rafting and volcanic inputs
510 are the only other viable mechanisms at Site U1313) because additional terrigenous
511 inputs would act to amplify our terrigenous record rather the biomarker record (ice
512 rafting control on biomarker flux is not documented even during the extreme Heinrich
513 events of the Late Pleistocene, Naafs et al. 2012). Furthermore, our records show that,
514 where present (low values from MIS G4), IRD accumulation rates are always higher
515 in glacials than in interglacials (sand-sized volcanic grains are extremely rare in Site
516 U1313 sediments throughout our study interval; Figure 5). Thus, there is no way to
517 explain amplification of the glacial jumps in the biomarker record (relative to the
518 terrigenous fraction) by invoking decreases in IRD and/or volcanics inputs while
519 a linear relation is maintained between biomarker and lithogenic dust. In other words,
520 our records point to the unequivocal existence of some mechanism that acts to
521 amplify the glacial jumps in the biomarker record relative to those in our terrigenous
522 record.

523 Amplification of the glacial biomarker signal from MIS G6, and particularly
524 during MIS 100 (Figure 10) points to increased efficiency of biomarker export/burial
525 (especially in n-alkan-1-ols) and/or major shifts in vegetation biomes relative to
526 preceding glacials. It seems an unlikely co-incidence that MIS 100 is the oldest glacial

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527 for which there exists convincing evidence from diverse proxy records for the
528 existence of a major Laurentide Ice Sheet (Bailey et al., 2010; 2013; Bringham-Grette
529 et al., 2013) extending well into the mid-latitudes (39° north based on the terrestrial
530 record of glacial tills; Balco and Rovey, 2010). We hypothesize that some
531 combination of a southward shift of boreal and temperate forest biomes across North
532 America, strengthening of wind-driven sand-blasting and perhaps precipitation-led
533 increase in woody plant cover (woody thickening) in arid regions south of the
534 Laurentide Ice Sheet front may be responsible for the amplified glacial jumps in the
535 biomarker records, especially the extreme signal seen in MIS 100. Our hypothesis
536 requires testing but is consistent with the interpreted response of the atmosphere and
537 vegetation to ice sheet advance well into the mid-latitudes during the Last Glacial
538 Maximum (LGM) (e.g., Clark & Pollard, 1998; Kutzbach et al., 1998; Clark et al.,
539 1999; Thompson and Anderson 2000; Huang et al., 2001; Prentice et al., 2011; Bragg
540 et al., 2013; Ullman et al., 2014).

541

542 *3.6. Eolian dust deposition in the North Atlantic Ocean during the warm Pliocene.*

543 We argue that the onset of clear glacial-interglacial cycles in sediment color is driven
544 by changes in terrigenous dust accumulation at Site U1313 and that these cycles
545 appear at least 800 kyr earlier than MIS 100 and well before significant iNHG
546 commenced around 2.72 Ma. In Figure 11 we assess how far back into the Pliocene
547 Epoch these signals extend by comparing sediment color reflectance and estimated
548 lithogenic dust flux from Site U1313 to published climate records for the entire Plio-
549 Pleistocene (to ~5.3 Ma, the base of LR04). The correspondence between sediment
550 color at Site U1313 and global climate change registered by benthic $\delta^{18}\text{O}$ is
551 remarkable. With the exception of one main interval of peak Pliocene warmth (4.3 to

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552 ~4.0 Ma; Seki et al., 2012) when the sediment color reflectance record shows high
553 values with little orbital structure and a minor contribution from lithogenic dust can
554 be inferred, we observe the Pleistocene pattern (L^* minima during glacials; maxima
555 during interglacials) at Site U1313 back to 5.3 Ma (the base of LR04, Fig. 11A).

556 In some respects, the signal of a minor lithogenic dust component during high
557 CO_2 warm Pliocene conditions is expected because climate model simulations (e.g.
558 Salzmann et al., 2008; 2013; Goldner et al., 2011) and paleo-data (e.g. Zarate and
559 Fasana, 1989; Thompson, 1991; Smith, 1994; Axelrod, 1997; Salzmann et al., 2008,
560 2009, 2013; Jimenez-Moreno et al., 2010) for the warm Pliocene, particularly for the
561 Mid-Piacenzian PRISM time-slab (Dowsett et al., 2012; Haywood et al., 2013),
562 indicate noticeably wetter than modern conditions in modern arid and semi-arid
563 regions, including the American Southwest. Yet in other respects our findings are
564 surprising because there is broad consensus among climate model predictions for the
565 future suggesting an increase in the expanse of arid to semi-arid mid-latitudes in a
566 warmer world, and that this transition should already be underway in North America
567 (e.g. Held and Soden, 2006; Seager et al., 2007; O’Gorman and Schneider, 2009).

568 Three main hypotheses have been suggested to explain the fundamental
569 discrepancy between climate model predictions for the next 50 to 100 years and the
570 model simulations of the warm Pliocene: (i) Differing boundary conditions, in
571 particular the effect on regional precipitation fields of a potentially markedly lower
572 elevation of the Pliocene Rocky Mountains (Wolf et al., 1997; Bonham et al., 2009)
573 prior to the mid Pliocene (Foster et al., 2010). (ii) Enhanced regional precipitation in
574 (southwest) North America relative to today in response to a warm eastern equatorial
575 Pacific (Fig. 11B) in an El Niño-prone world (Goldner et al., 2011). (iii) Fundamental
576 differences in the climate signal being modeled (equilibrium condition Pliocene

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577 climates incorporate both short and long-term feedbacks associated with climate
578 sensitivity while predictions for the non-equilibrium condition ‘climate transient’ of
579 the coming 50 years necessarily incorporate only fast or Charney feedbacks
580 (Salzmann et al., 2009)).

581 Each of these hypotheses makes different predictions for the timing of the
582 onset of source aridification and dust generation spatially through Pliocene time,
583 thereby presenting a means to test their validity. For example, the disappearance of
584 summer wet flora in North American terrestrial records that span the Miocene-
585 Pliocene boundary on both sides of the Cascades and Sierra Nevada mountains
586 suggests that aridification of the American West through the Mio-Pliocene is unlikely
587 to be related to a rain shadow effect due to mountain uplift (Lyle et al., 2008).
588 Similarly, based on global terrestrial vegetation reconstructions, the picture of a
589 wetter-than present warm Pliocene appears to be too extensive (Salzmann et al., 2009;
590 2013) to support the suggested role of North American mountain orography. But
591 while terrestrial records of precipitation balance provide powerful insights into
592 Pliocene climate (Salzmann et al., 2013), they are, by their nature, discontinuous in
593 coverage and often suffer from age control limitations. Plio-Pleistocene data coverage
594 for mid-latitude North America, including for the core of the present-day arid
595 American Southwest, is extremely poor because of the lack of lacustrine deposits
596 generally and Pleistocene glacial erosion in the north (Salzmann et al., 2009; 2013).
597 The secular signal in the Site U1313 record is broadly consistent with the
598 hypothesized importance of warm sea surface temperatures in the Pliocene eastern
599 equatorial Pacific (Fig. 11). Yet, many differences between early Pliocene and
600 present-day climates of parts of Africa, Asia, and Australia do not resemble the
601 anomalies associated with canonical El Niño teleconnections (Cane and Molnar,

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2007). Alongside model-based evaluation of the influence of fast versus slow
feedbacks on precipitation balance and proxy reconstructions of the hydrological
cycle, improved records of Pliocene dust deposition in well-dated marine sites
recovered downwind from known Quaternary dust source regions will provide a
valuable means to help understand the climatic response to sustained global warmth in
the recent geological past.

608

4. Conclusions

We present Plio-Pleistocene records of sediment color, %CaCO₃, foraminifer
fragmentation, benthic $\delta^{13}\text{C}$, coarse lithic counts and the radiogenic isotope (Nd, Sr,
Pb) composition of terrigenous sediment component from IODP Site U1313. We
demonstrate that glacial-interglacial cycles in sediment color are unambiguously
correlated to benthic $\delta^{18}\text{O}$ back to at least 3.3 Ma, and represent changes in sediment
%CaCO₃. Our new records of terrigenous and carbonate sediment accumulation rates,
foraminifera fragmentation and benthic $\delta^{13}\text{C}$ show that these cycles are driven by
enhanced glacial fluxes of terrigenous material and not glacial dissolution of
carbonate material as previously interpreted.

On the basis of our radiogenic isotope data, we rule out a high-latitude origin
for the terrigenous sediment component deposited at Site U1313 during our study
interval and suggest that eolian dust sourced from mid latitude North America
dominates clastic sediment deposition at this site during the Pliocene. This finding is
consistent with previously published inferences on the provenance of an n-alkane
biomarker proxy for dust inputs to our study site. Together with the biomarker
records, our lithogenic data sets demonstrate that North America shifted abruptly to a
distinctly more modern cold and arid glacial regime from MIS G6 with the

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627 development of a Laurentide ice sheet extending well into the mid-latitudes by MIS
628 100. Yet the relation between the biomarker and lithogenic component of dust
629 accumulation at Site U1313 is distinctly non-linear. Both records show a jump in
630 glacial rates of accumulation from ~2.7 Ma onwards (during MIS G6) but the
631 amplitude of this signal is about 3 to 8 times greater for biomarkers than for lithogenic
632 dust and particularly extreme during MIS 100 (2.52 Ma).

633 The development of significant continental ice in the northern hemisphere
634 during glacials from MIS G6 onwards undoubtedly had a profound impact on dust
635 generation on North America. Our analysis, however, suggests that glacial grinding
636 and transport of fine grained sediments to mid latitude outwash plains is not the
637 fundamental mechanism controlling the magnitude of the flux of higher plant leaf
638 waxes from North America to Site U1313 during iNHG. We hypothesize that some
639 combination of latitudinal biome shift, strengthening of sand-blasting south of North
640 American ice sheet front and perhaps precipitation-led woody thickening of arid
641 regions in response to ice sheet advance towards the mid-latitudes may be responsible
642 for the non-linearity observed. The secular pattern of change in the North Atlantic
643 record indicates that there existed a minor lithogenic dust component at our study site
644 during high-CO₂ peak Pliocene warm conditions (in contrast to climate model
645 predictions for the future suggesting an increase in the expanse of arid to semi-arid
646 zones in a warmer world). At least part of the discrepancy between climate model
647 predictions for enhanced aridity of the mid latitudes over next 50 to 100 years and
648 geologic observations for a warm wet Pliocene is likely attributable to fundamental
649 differences in the climate signal being observed for the Pliocene versus that being
650 modeled for future decades (equilibrium condition Pliocene climates versus transient
651 non-equilibrium model predictions for the future). The form of secular change shown,

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652 however, is broadly consistent with the hypothesized importance of warm sea surface
653 temperatures in the eastern equatorial Pacific during the Early Pliocene in bringing
654 about wetter-than-modern conditions in mid-latitude North America.

655

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1162

1163 **Figure captions**

1164 **Figure 1.** Lithostratigraphic cycles in North Atlantic deep-sea sediments of Plio-
1165 Pleistocene age in multiple drill sites as revealed by published physical property
1166 records. Sites arranged (from top to bottom) in order of increasing water depth. Note
1167 the existence of clear rhythmic cycles significantly earlier than MIS 100, the inferred
1168 glacial for the onset of basin-wide ice rafting. The data presented in the bottom panel
1169 (A) were originally compiled by Ruddiman et al. (1987, their Figure 3), although we
1170 substitute their record from DSDP Site 607 with our higher resolution proxy record
1171 from IODP Site U1313 (See Section 2.4 for methods). Those authors concluded that
1172 %CaCO₃ variability at pelagic DSDP Sites 607 (U1313) and 609, but not at the
1173 shallower DSDP Site 552, prior to Gauss/Matuyama boundary time was attributable
1174 to sea floor CaCO₃ dissolution, a consequence of the influence of corrosive poorly
1175 ventilated glacial intermediate waters in the North Atlantic. Top panel (B) shows data
1176 generated from additional shallow sites drilled after the Ruddiman et al., (1987) study
1177 (pelagic Site 982 (Shipboard Scientific Party, 1996), drift Site 980/981 (Ortiz et al.,

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1178 1999)). Note that the timing of the initiation of marked lithological cycles in the North
1179 Atlantic drill sites is not a simple function of water depth suggesting that CaCO_3
1180 dissolution is not the principle origin of these cycles (see text). The horizontal
1181 black/white bars in each panel denote paleomagnetic (sub)chronozone boundaries
1182 (Cande and Kent, 1995): B = Brunhes, M = Matuyama, G = Gauss, K = Keana, Ma =
1183 Mammoth and Gil = Gilbert.

1184

1185 **Figure 2.** North Atlantic region showing location of IODP Site U1313 relative to
1186 other drill sites referred to in the text (**A**) and mean April to September (the ‘dust
1187 season’; Prospero et al., 2002) surface wind vectors (**B**; image source
1188 <http://www.esrl.noaa.gov/psd/> (Kalnay et al., 1996)). Also shown in (**A**) is the last
1189 glacial maximum IRD-belt (stippled area) of Ruddiman (1977), relevant principal
1190 surface-ocean current systems (adapted from Kleiven et al., 2002) and average
1191 radiogenic isotope composition of potential source regions of terrigenous sediments
1192 deposited at Site U1313 (based on data shown in Figure 4).

1193

1194 **Figure 3.** Relationship between IODP Site U1313 sediment color (L^*) and calcium
1195 carbonate ($\% \text{CaCO}_3$) content. Global benthic oxygen isotope stack for the Plio-
1196 Pleistocene, the LR04 (Lisiecki and Raymo, 2005) and published benthic oxygen
1197 isotope data for IODP Site U1313 (**A**); discrete $\% \text{CaCO}_3$ measurements for late
1198 Pleistocene (**B**, black circles, Stein et al., (2009), $n = 151$) and late Pliocene and
1199 earliest Pleistocene (**C**, red circles, this study, $n = 193$) and our high resolution
1200 estimate of sediment $\% \text{CaCO}_3$ (against meters composite depth, mcd) based on least
1201 squares linear regression of L^* (5-point, 10 cm, moving average) onto discrete
1202 $\% \text{CaCO}_3$ measurements (**D**). Data corresponding to North Atlantic Hudson Strait
1203 Heinrich-like events (vertical grey bars in **B** labelled HE), for which the relationship

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1204 between L^* and $\%CaCO_3$ breaks down, are excluded from our least square regression.
1205 We identified Heinrich Layers at Site U1313 following Stein et al. (2009), based on
1206 their high (>500 cps) x-ray diffraction-derived dolomite concentrations. The
1207 horizontal black/white bars in each panel denote paleomagnetic (sub)chronozone
1208 boundaries (Cande and Kent, 1995): B = Brunhes, M = Matuyama, G = Gauss, K =
1209 Keana, Ma = Mammoth, Gil = Gilbert, C = Cochiti, N = Nunivak and S = Sidufjall.
1210 Depth range of chronozone boundaries shown in (B) and (C) based on shipboard
1211 measurements (Expedition 306 Scientists, 2006).

1212

1213 **Figure 4.** Characterisation of likely sources of terrigenous sediment to Site U1313 in
1214 Nd-Sr (A) and Pb-Pb (B & C) spaces. These fields are based on modern bedrock,
1215 loess, river sediment and aerosol data, and modern to LGM ice sheet/dust source
1216 proximal sediment core data. Potential sources constitute high-latitude material from
1217 Greenland and Northern Canada (the Canadian Province, Blue), volcanic material
1218 from Eastern Greenland and Iceland (Red, together with the Canadian Province this
1219 represents the most likely source of ice rafted material), mid-latitude material from
1220 Europe and the Gulf of St. Lawrence (purple) that is unlikely to be a significant
1221 source of ice-rafted material prior to significant northern hemisphere glaciation and
1222 potential aeolian sources from North America and the Sahara (green and yellow
1223 bubbles, respectively). Fields based on data from (also see Supplementary
1224 Information, Figures S1–S5): Abouchami and Zabel. (2003), Aleinkoff et al. (1999),
1225 Aleinkoff et al. (2008), Asmerom and Jakobsen (1993), Bernstein et al., (1998),
1226 Biscaye et al. (1997), Cohen and O’Nions (1982), Cole et al. (2009), Farmer et al.
1227 (2003), Goldstein and Jakobsen (1988), Grousset et al. (1988), Grousset et al. (2001),
1228 Hansen and Nielsen (1999), Juteau et al. (1986), Kokfelt et al. (1983), Kokfelt et al.

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1229 (2006), Millot et al. (2004), Revel et al. (1996).
1230
1231 **Figure 5.** Paleoceanographic records from IODP Site U1313 for the late Pliocene and
1232 earliest Pleistocene: **(A)** Foraminifera fragments as a percentage of total foraminifera
1233 plus fragments observed in the >150 μ m fraction (Ivanova et al., 2003). Overall, the
1234 carbonate material from this site is exceptionally well preserved. Modest increases in
1235 fragmentation are observed, however, during glacial periods from ca. 2.52 Ma
1236 onwards, demonstrating that more corrosive conditions existed at this site during
1237 glacials MIS 100, 98 and 96. For reference, we have included a line that approximates
1238 $\Delta(\text{CO}_3^{2-}) = 0$ in terms of percentage fragmentation based on Le and Shackleton
1239 (1992); **(B)** Benthic foraminiferal $\delta^{13}\text{C}$, measured on *Cibicidoides wuellerstorfi* (this
1240 study); **(C)** Alkenone mass accumulation rates, a productivity proxy (Naafs et al.,
1241 2010); **(D)** Mass accumulation rates, MAR, of n-alkanes and C26-alkan-1-ol, aeolian
1242 derived biomarkers (Naafs et al., 2012); **(E)** Calculated MAR of terrigenous material
1243 (this study); **(F)** Concentration of ice rafted coarse lithics (Ice rafted detritus, IRD,
1244 >150 μ m, excluding volcanics, which are only ever present in trace numbers).
1245 Transparent grey box shows the range of peak glacial values estimated for high-
1246 latitude North Atlantic Ocean DSDP Site 611 between MIS G6-100 (~2.72-2.5 Ma;
1247 Bailey et al., 2013). Overall, coarse lithic content of iNHG sediments at Site U1313 is
1248 extremely low (<50 grains g^{-1}) and prior to MIS 100 never higher than 5 grains g^{-1}
1249 (contrast with extremely high concentrations of 1500-5000 g^{-1} at Site 611); **(G)**
1250 Benthic $\delta^{18}\text{O}$ (Bolton et al., 2010) and L* derived % CaCO_3 (this study), a remarkable
1251 correlation is seen. All data plotted on age model of Bolton et al. (2010). All MAR
1252 data shown estimated (as $\text{MAR} = \text{component abundance} \times \text{linear sedimentation rate} \times$
1253 dry bulk density) using sedimentation rates based on the age model of Bolton et al.

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1254 (2010) and dry-bulk densities from shipboard determined GRAPE wet-bulk density
1255 data following the approach of Maslin et al. (1995). MARs shown in (C) and (D)
1256 recalculated on this basis using published datasets, but do not differ appreciably from
1257 original fluxes reported by Naafs et al. (2012). The horizontal black/white bars at the
1258 base of the figure denote paleomagnetic chronozone boundaries (Cande and Kent,
1259 1995): M = Matuyama, G = Gauss, K = Keana and Ma = Mammoth.

1260

1261 **Figure 6.** Pb (A & B) and Nd-Sr (C) isotope composition of Plio-Pleistocene IODP
1262 Site U1313 bulk terrigenous sediments and range of radiogenic isotope values for
1263 potential terrigenous sources (as compiled in this study, see supplementary
1264 information, Figures S1–S5). Data from interglacial (triangles) and glacial (circles)
1265 samples are highlighted. Data uncertainty (at 2σ) is plotted, but usually smaller than
1266 symbols shown.

1267

1268 **Figure 7.** Time series of the radiogenic isotope composition of bulk terrigenous
1269 sediments deposited at IODP Site U1313: $^{87}\text{Sr}/^{86}\text{Sr}$ (A) ϵNd (B), $^{206}\text{Pb}/^{204}\text{Pb}$ (C).
1270 Benthic $\delta^{18}\text{O}$ stratigraphy for Site U1313 (Bolton et al., 2010) shown for reference.
1271 Grey dashed lines highlight relationship between data in (A-C) and glacial and
1272 interglacial marine isotope stages shown in (D). Data uncertainty (at 2σ) in A-C is
1273 smaller than symbols used. The radiogenic isotope composition of source regions
1274 shown on right hand side of figure (median – black line, 66th percentile – box, 95th
1275 percentile – “whisker”, outlying data-points marked as small crosses) are determined
1276 from data shown in Figure 4. Horizontal green lines denote the median of North
1277 American loess measurements, concluded to be the dominant source of Site U1313
1278 terrigenous sediments (see main text). For comparison, Nd and Sr isotope

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1279 measurements from last glacial maximum ice rafting events at nearby drill cores
1280 (Sites SU90-08 and SU90-09 (Revel et al., 1996; Grousset et al., 2001)) are shown in
1281 (A) and (B). These data reveal extensive variability during the last glacial cycle (from
1282 -5.8 to -40.9 ϵNd , 0.72904 to 0.71662 $^{87}\text{Sr}/^{86}\text{Sr}$) and demonstrate that the radiogenic
1283 isotope systems studied are sensitive to large ice rafted debris inputs when present. Ice
1284 rafted debris is first observed at Site U1313 during MIS G6 (labelled in D). The
1285 horizontal black/white bars at the base of the figure denote paleomagnetic chronozone
1286 boundaries (Cande and Kent, 1995): M = Matuyama, G = Gauss, K = Keana and Ma
1287 = Mammoth.

1288

1289 **Figure 8.** Wavelet analysis of North Atlantic ODP Site 659 terrigenous accumulation
1290 rate (A) (Tiedemann et al., 1994), and Site U1313 terrigenous accumulation rate (B)
1291 and benthic oxygen isotopes (Bolton et al., 2010) (C). Wavelet spectra estimated
1292 following (Torrence and Compo, 1998). Solid black lines in each panel enclose
1293 regions of >95% confidence, based on a red-noise model (Torrence and Compo,
1294 1998). Within light shaded areas of panel B and C confident interpretation cannot be
1295 drawn due to edge effects (Torrence and Compo, 1998). These effects are not visible
1296 in (A) because we show only a central portion of a 5.3 Ma record analysed.
1297 Horizontal dashed grey lines on each panel labeled with 19, 23, 41 and 100 pick out
1298 dominant periodicities of orbital cycles. The dominant 20 ka, precessional cyclicity
1299 seen in Saharan dust inputs to Site 659 is not found in the mass accumulation rate of
1300 terrigenous sediment at Site U1313. The age model used for Site 659 is based on that
1301 published in Tiedemann et al. (1994). Re-analysis of the Site 659 terrigenous
1302 accumulation rate record following retuning of its benthic $\delta^{18}\text{O}$ stratigraphy to the
1303 LR04 stack (Lisiecki and Raymo, 2005) does not remove the strong 20 ka

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1304 precessional cyclicity shown in panel A (see Supplementary Information).

1305

1306 **Figure 9.** Cross plots of late Pliocene and earliest Pleistocene (3.33–2.41 Ma) IODP
1307 Site U1313 paleoceanographic and paleoclimate proxies: **(A)** Non-linear relationship
1308 between accumulation rates of terrigenous sediment (this study, interpreted as
1309 dominantly eolian dust) and dust-derived organic biomarkers (Naafs et al., 2012); **(B)**
1310 Non-linear relationship between global climate as recorded by benthic oxygen
1311 isotopes at Site U1313 (Bolton et al., 2010) and dust-derived organic biomarkers
1312 (Naafs et al., 2012); **(C)** Linear relationship between inferred eolian dust
1313 accumulation rates and dust-derived biomarker accumulation rates at Southern Ocean
1314 ODP Site 1090 (42°54.8'S, 8°54.0'E (Martinez-Garcia et al., 2011)). **(D)** Linear
1315 relationship between Site U1313 benthic oxygen isotopes (Bolton et al., 2010) and
1316 accumulation rates of terrigenous sediment (interpreted here as dominantly eolian
1317 dust). Note, we only plot data older than 2.41 Ma for Site U1313 when we can be
1318 confident that the bulk terrigenous sediment component at this site is dominated by
1319 eolian dust. Site 1090 data represent the last 4 Ma of aeolian dust deposition at this
1320 site. Biomarker accumulation rates used in this figure come from those plotted in
1321 Figure 4. Cross-plots in (A), (B) and (D) generated following linear interpolation of
1322 the terrigenous mass accumulation rate record to match the relatively lower resolution
1323 of the biomarker and benthic $\delta^{18}\text{O}$ data.

1324

1325 **Figure 10.** Time series of the ratio of accumulation rates of C26-alkan-1-ol and n-
1326 Alkane (Naafs et al., 2012) to terrigenous sediments at Site U1313. Both ratio time
1327 series are normalized to the average ratio observed for the Piacenzian PRISM time-
1328 slab (defined as 3.025-3.264 Ma). Biomarker accumulation rates used are the same as

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1329 those shown in Figure 5. Our higher-resolution record of terrigenous sediment
1330 accumulation rate is linearly interpolated to match the resolution of the biomarker
1331 records. The Site U1313 oxygen isotope stratigraphy (Bolton et al., 2010) is shown
1332 for reference, with key glacial marine isotope stages (M2, G6 and 100) labelled. Note,
1333 during glacial periods biomarker accumulation rates are enhanced relative to
1334 accumulation rates of bulk terrigenous material. The horizontal black/white bars at the
1335 base of the figure denote paleomagnetic chronozone boundaries (Cande and Kent,
1336 1995): M = Matuyama, G = Gauss, K = Keana and Ma = Mammoth.

1337

1338 **Figure 11.** The relationship between Site U1313 sediment lightness (L^*) and globally
1339 representative benthic $\delta^{18}\text{O}$, the LR04 (Lisiecki and Raymo, 2005) (A) and proxy
1340 indicators of the evolution of eastern equatorial Pacific (EEP) sea-surface temperature
1341 (B, based on alkenones from ODP Site 846 (Lawrence et al., 2006)) and sub-surface
1342 temperature (C, based on Mg/Ca ratios in foraminifer from ODP Sites 848, 849 and
1343 853 (Ford et al., 2012); ODP Site 1241 (Steph et al., 2006)). Horizontal dashed blue
1344 line in (B) corresponds to Holocene sea-surface temperature average for ODP Site
1345 846 (Lawrence et al., 2006). During the early Pliocene the mid latitudes of North
1346 America were wetter and warmer than present (Goldner et al, 2011). Note the warm
1347 temperatures of the EEP Ocean (associated with small zonal equatorial SST gradients
1348 (Ford et al., 2012), a state referred to as permanent El Niño-like (implies nothing
1349 about interannual variability). It is hypothesised that the development of the EEP cold
1350 tongue at this time and a subsequent poleward shift in the Pacific jet stream led to the
1351 aridification of North America (Goldner et al., 2011). Note that L^* at Site U1313, a
1352 proxy for sediment eolian content is unambiguously correlated to global climate
1353 (LR04) back to 3.3 Ma and intermittantly so probably back to 5.3 Ma (base of LR04),

North Atlantic Color Cycles

1354 the notable exception being ~4.3 to 4 Ma (see main text). For sediments older than 3.3
1355 Ma, our manual graphical correlation of Site U1313 and LR04 is based on tuning
1356 between constraints provided by shipboard determination of depths to
1357 paleomagnetostratigraphic boundaries (Expedition 306 Scientists, 2006). Age model
1358 control for ~2.4-3.3 Ma and <2.4 Ma, respectively, come from Bolton et al. (2010)
1359 and Naafs et al. (2012) and Expedition 306 Scientists (2006). The horizontal
1360 black/white bars at the top and base of the figure denote paleomagnetic chronozone
1361 boundaries (Cande and Kent, 1995): B = Brunhes, M = Matuyama, G = Gauss, K =
1362 Keana, Ma = Mammoth, Gil = Gilbert, C = Cochiti, N = Nunivak, S = Sidufjall and T
1363 = Thvera.

Figure 1

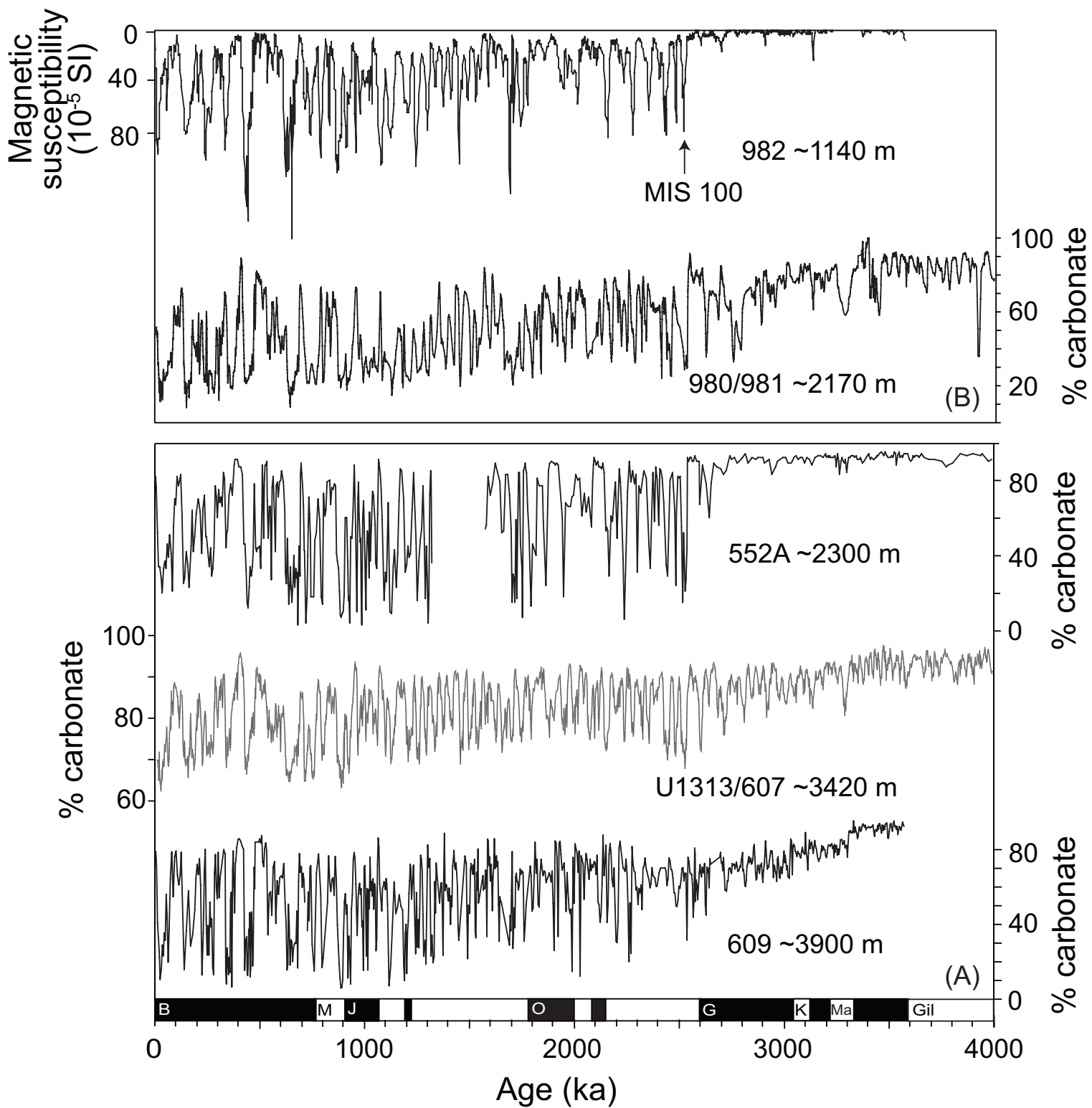


Figure 2

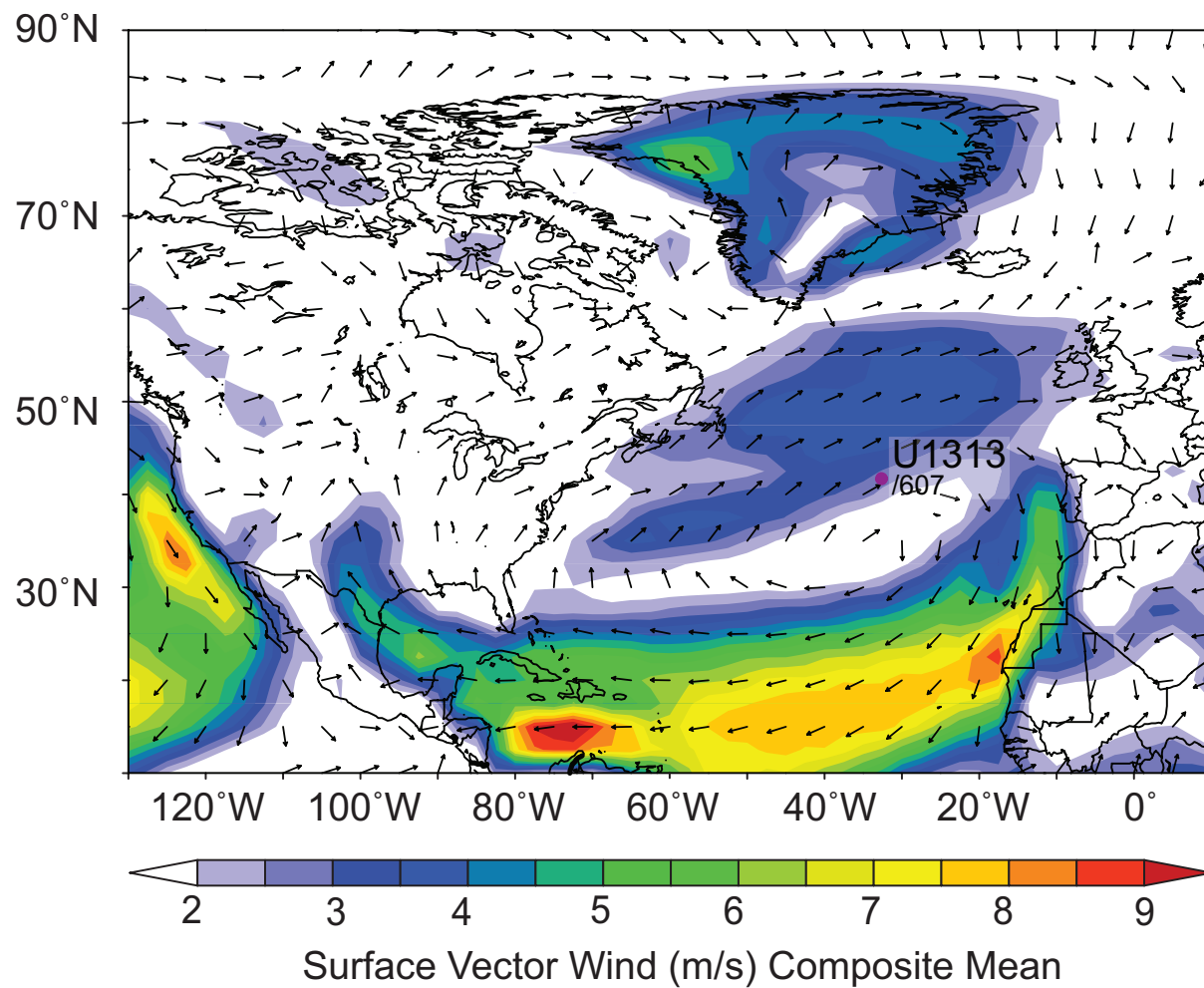
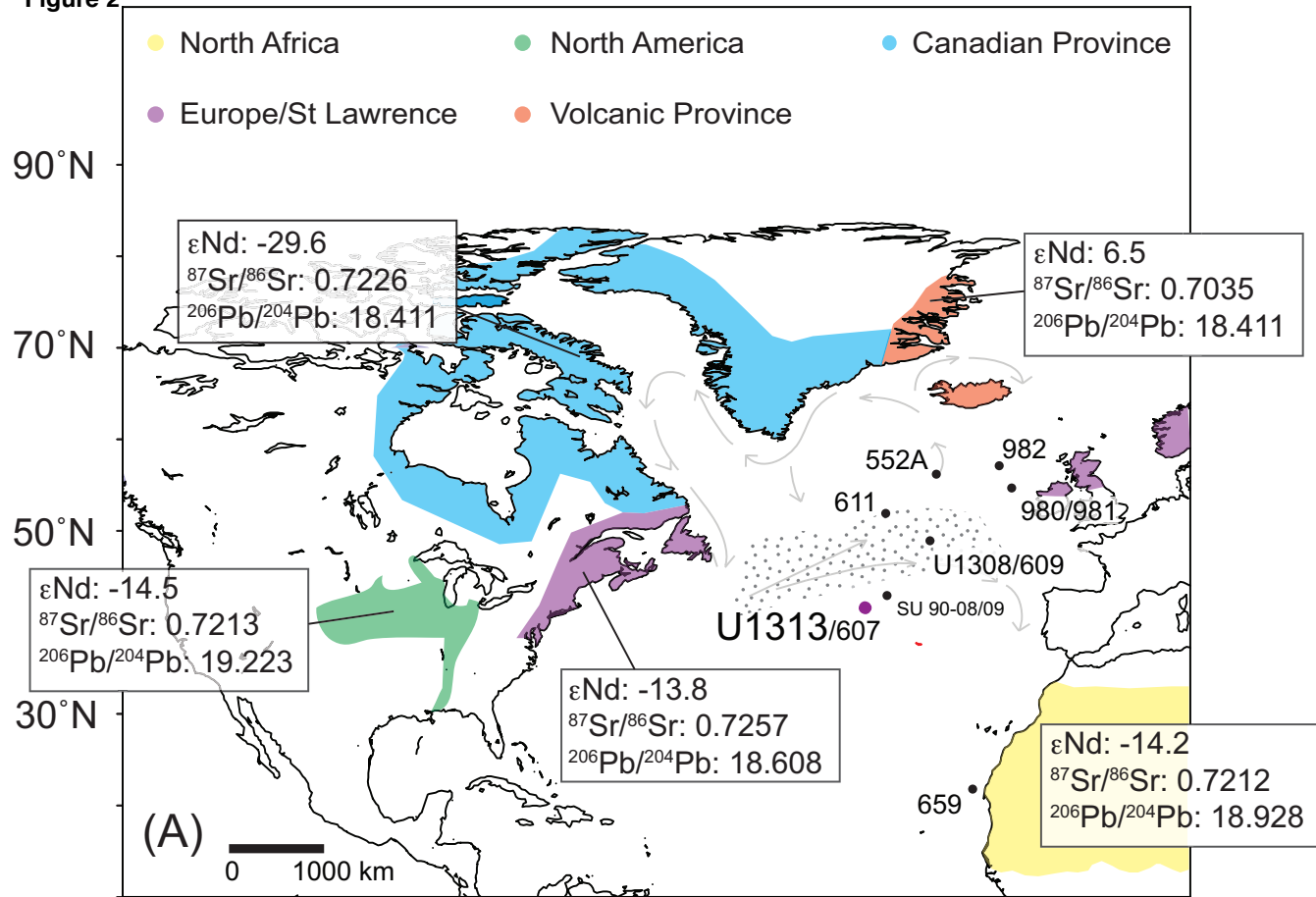


Figure 3

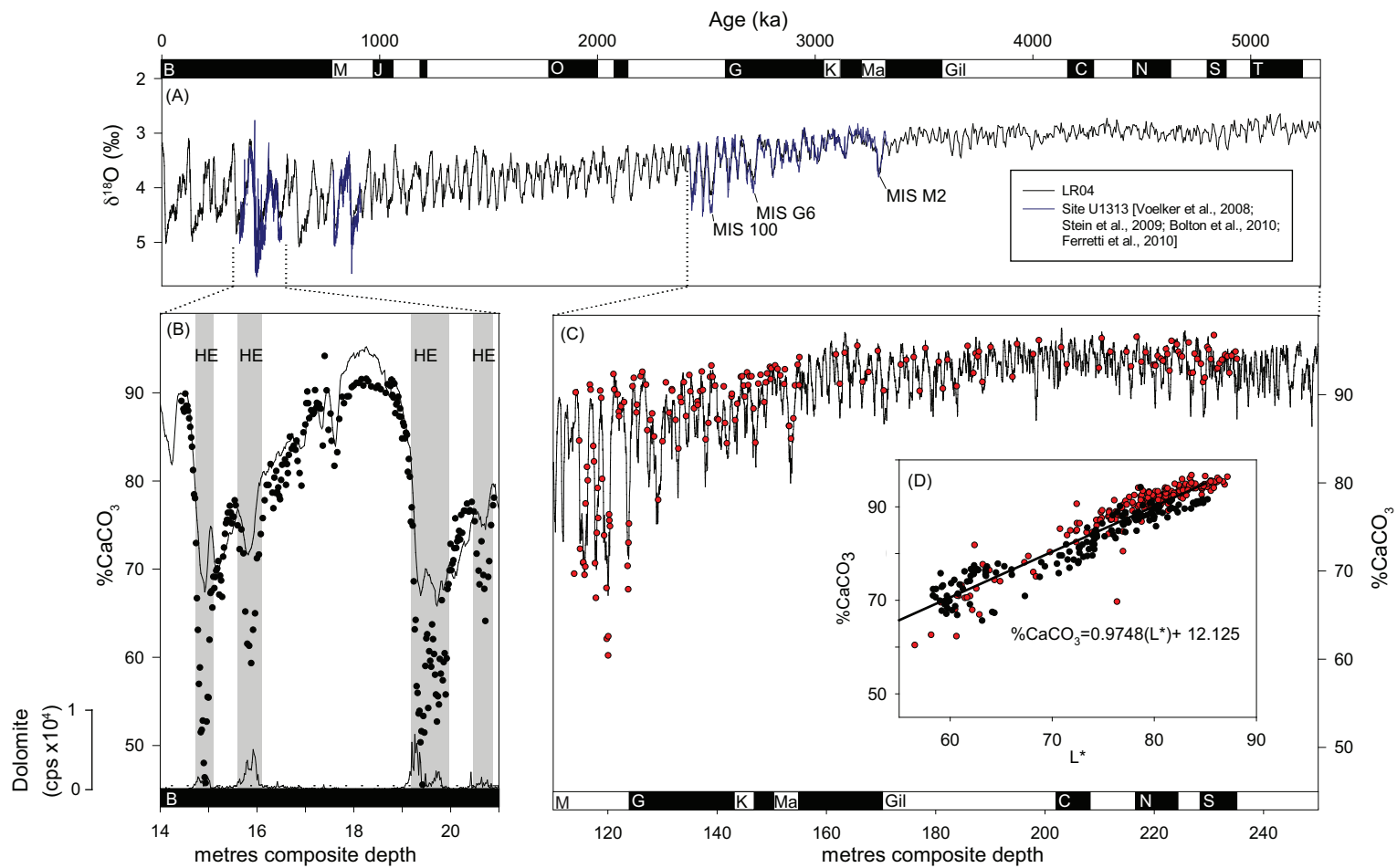


Figure 5

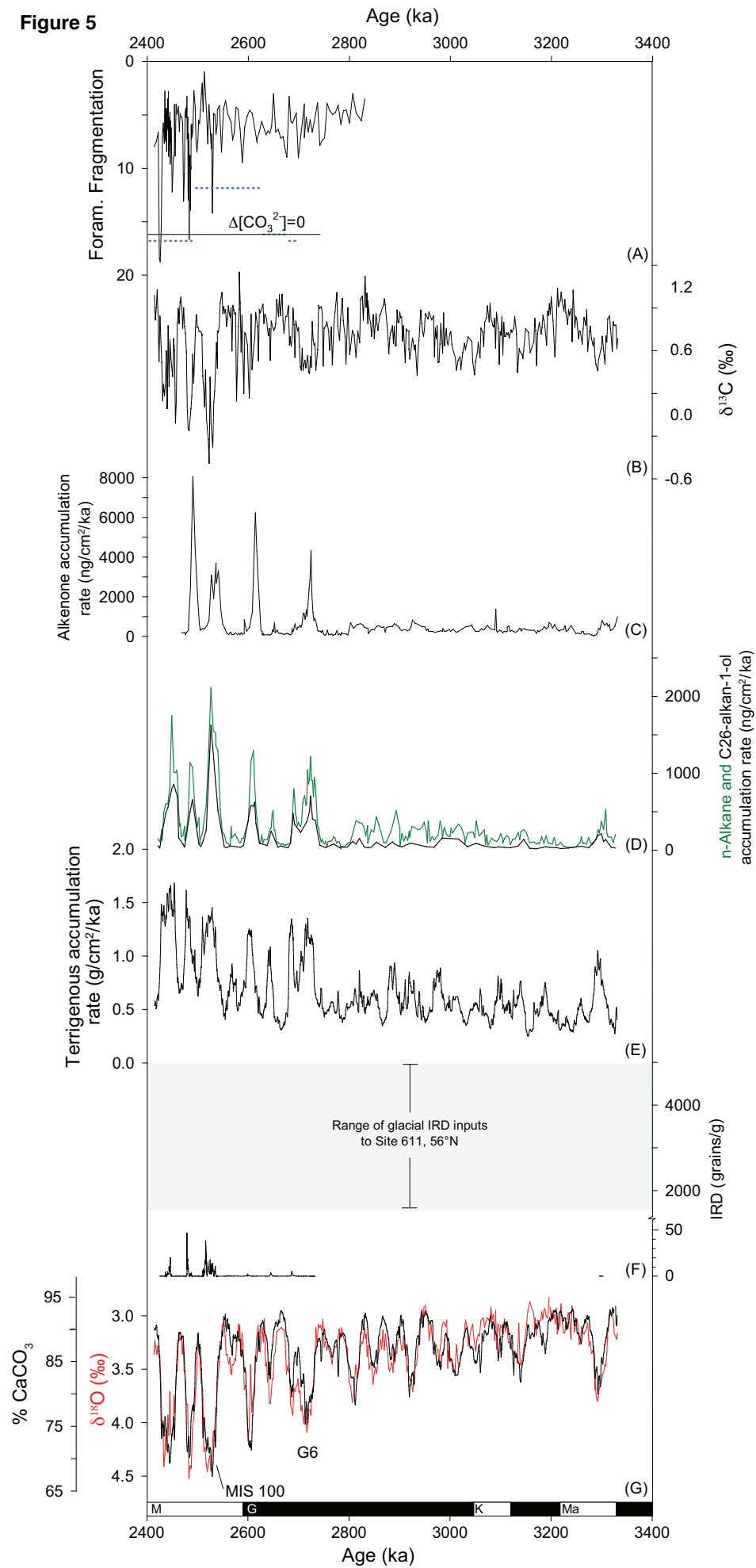


Figure 4

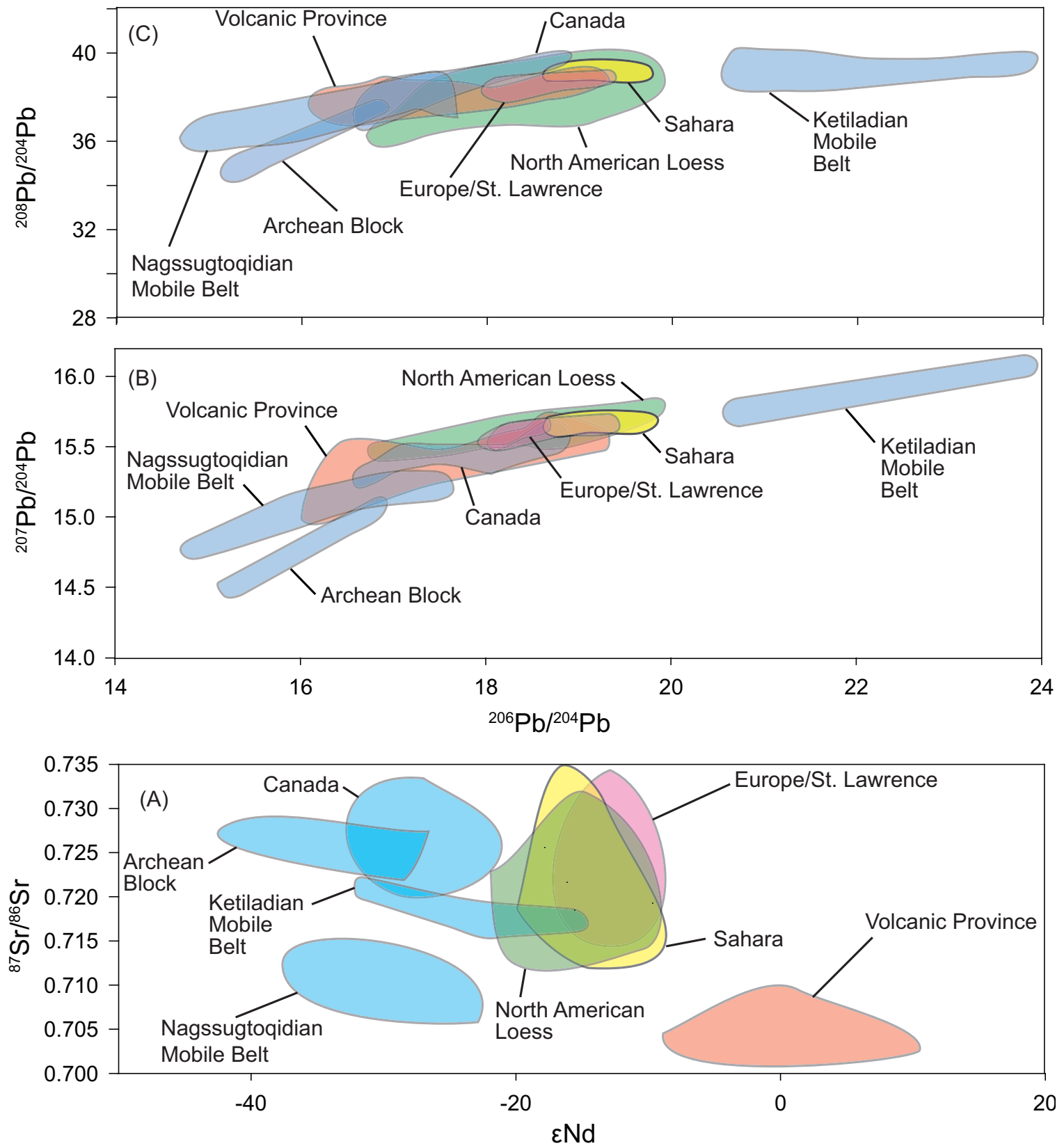


Figure 6

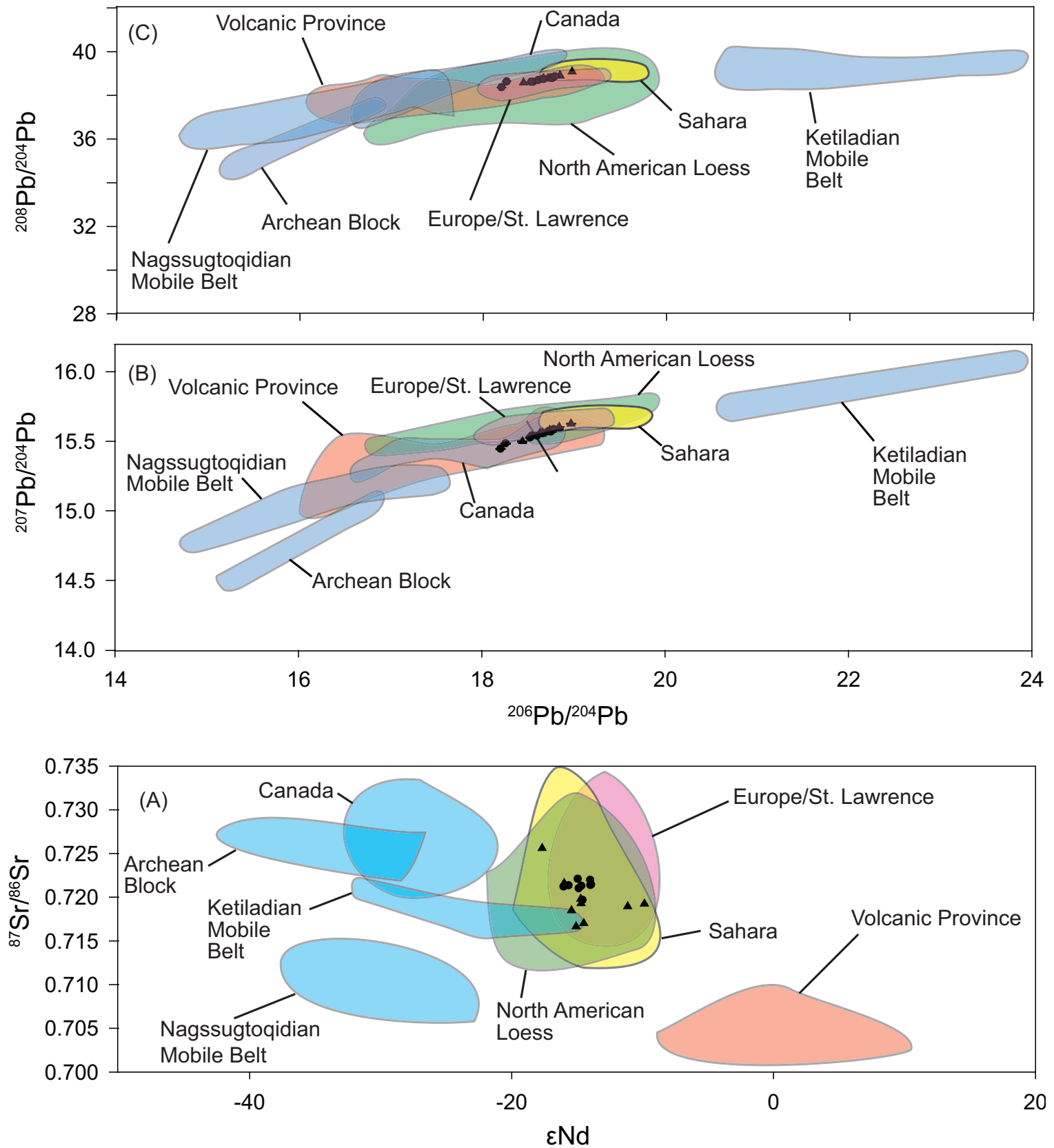


Figure 7

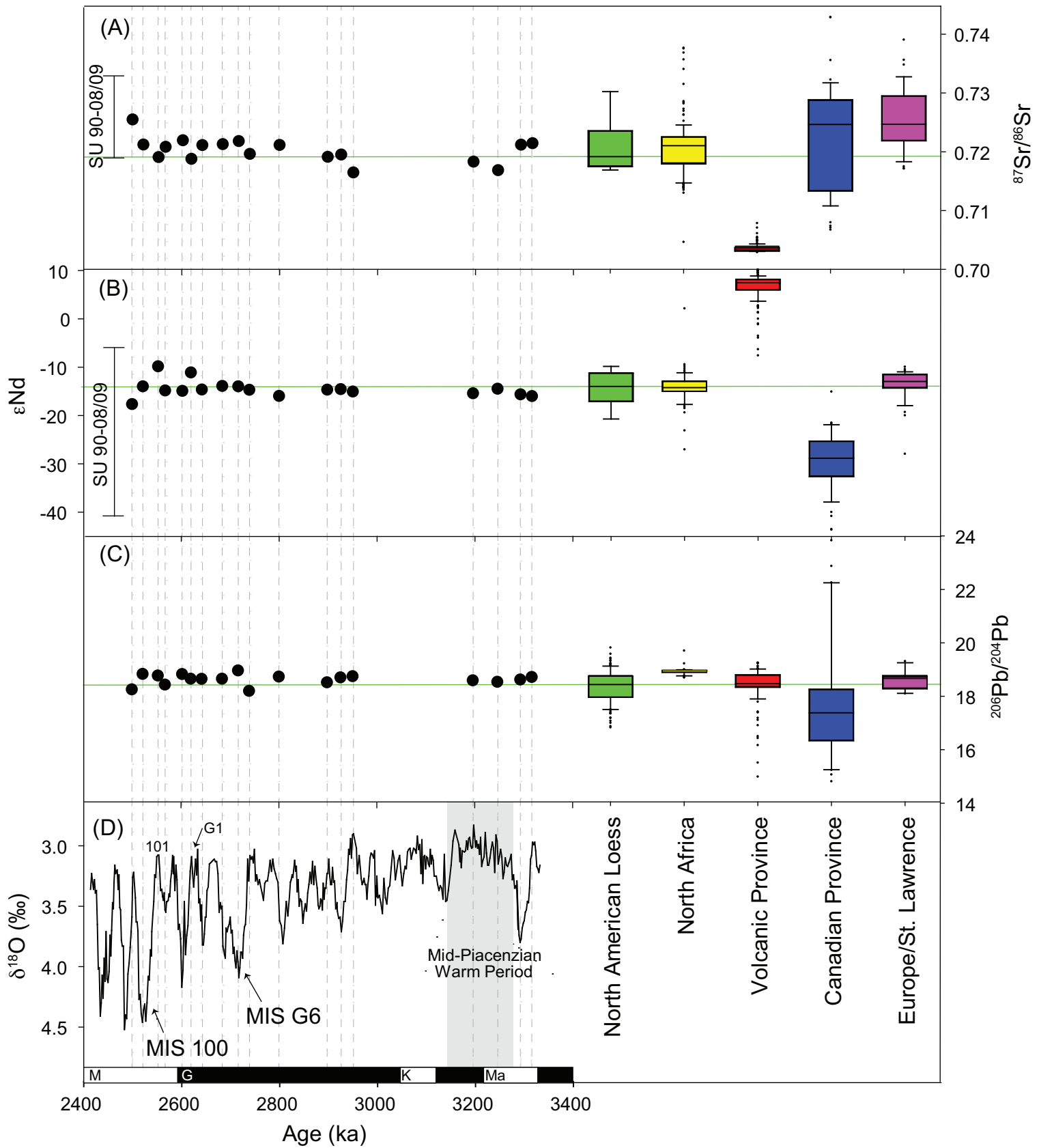


Figure 8

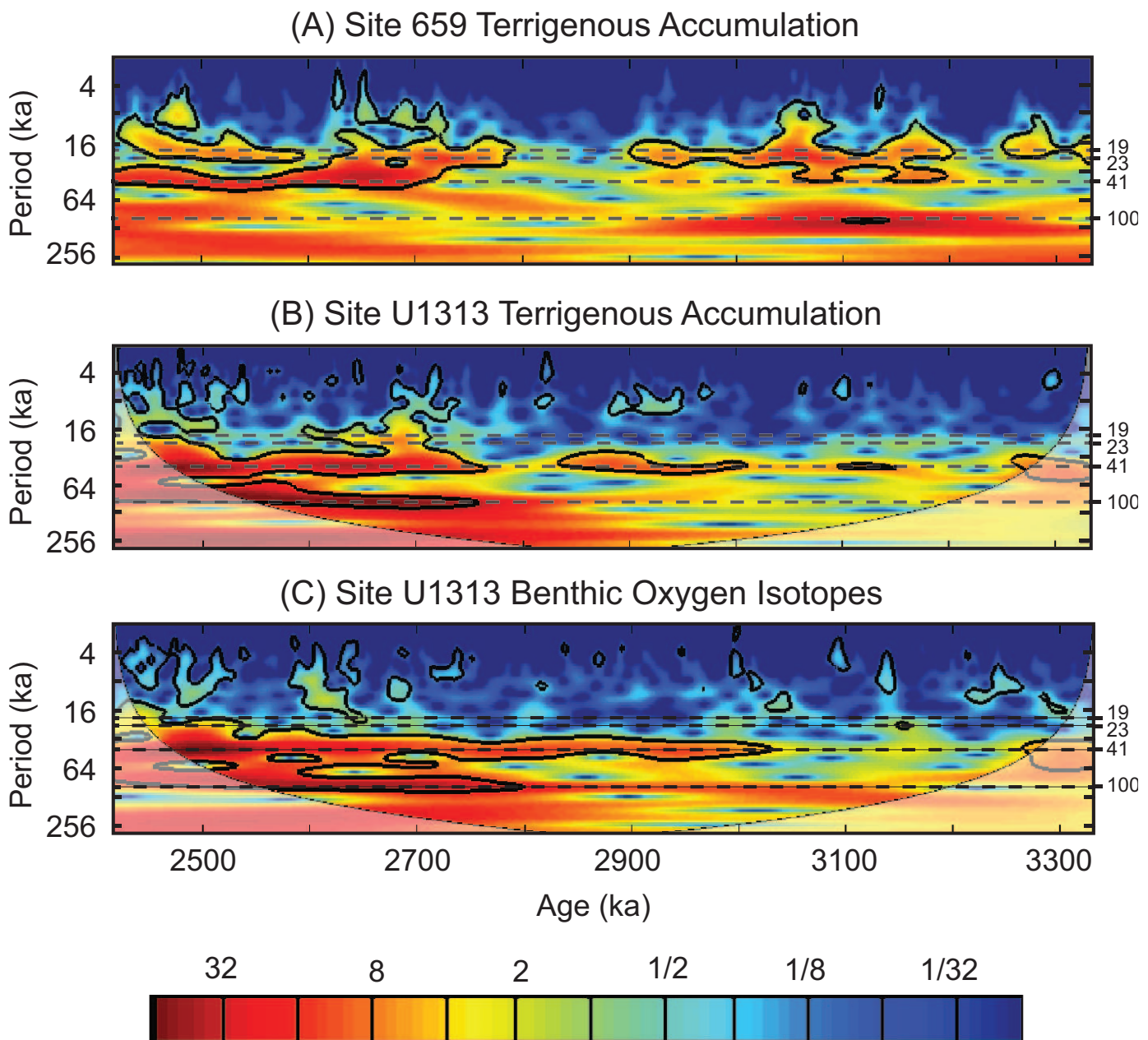


Figure 9

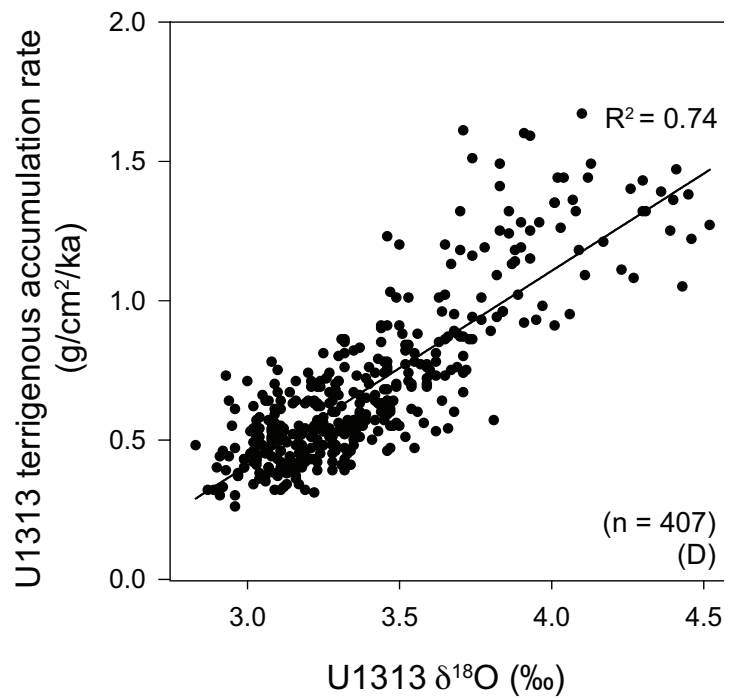
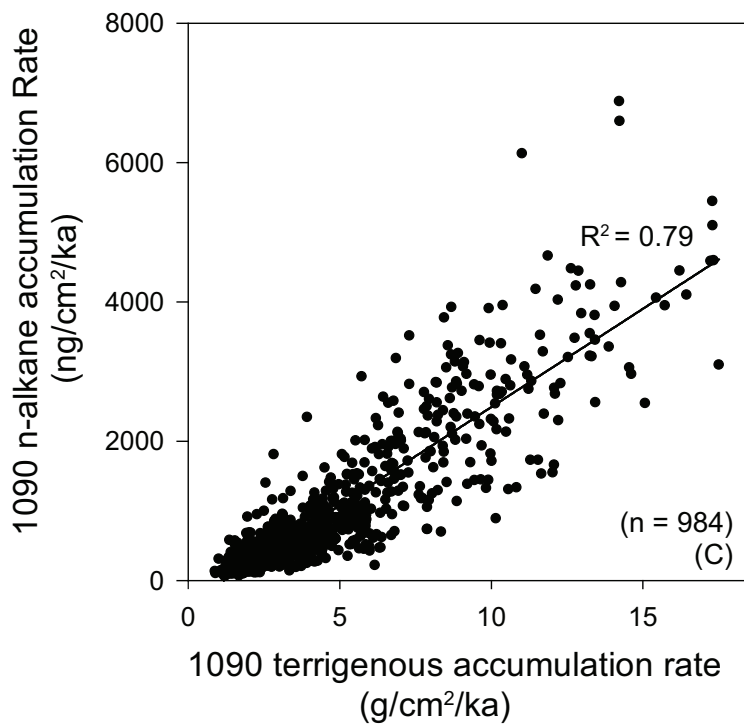
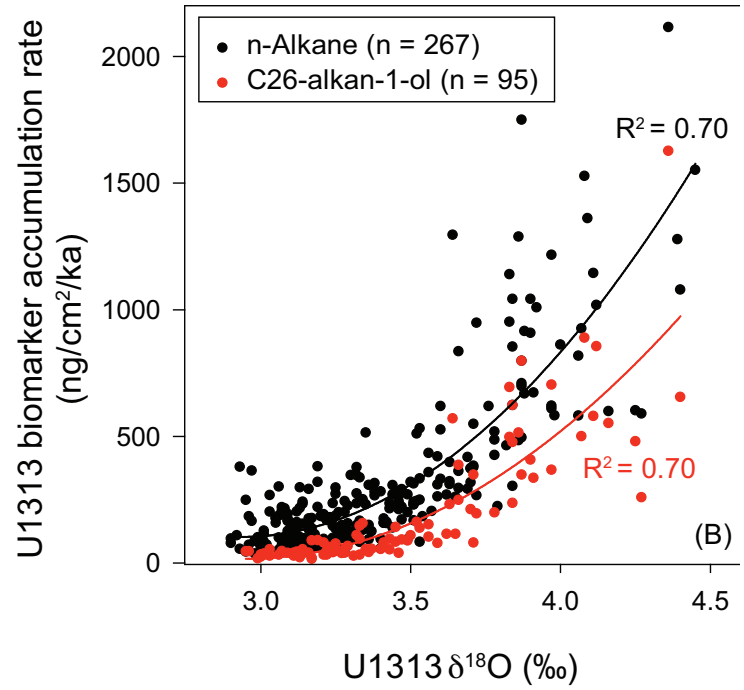
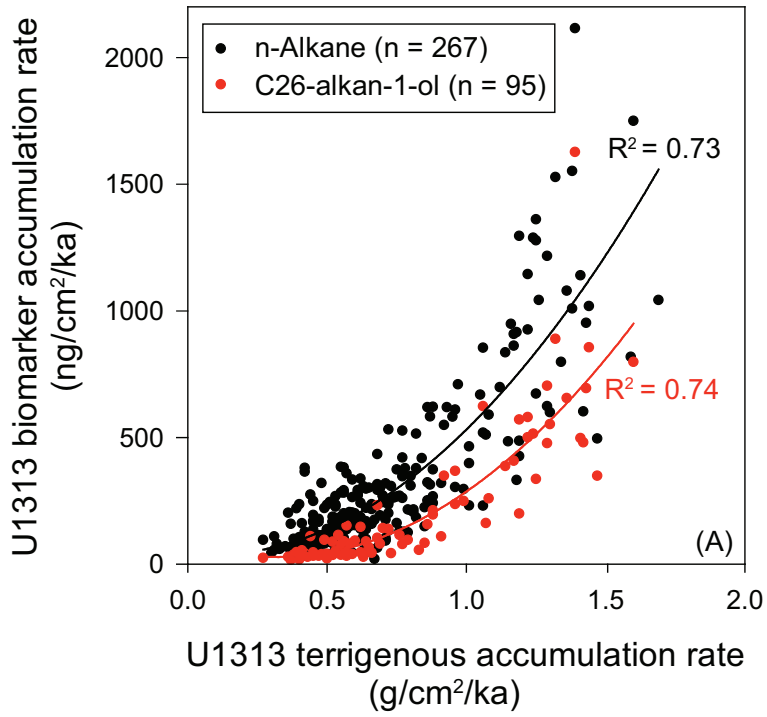


Figure 10

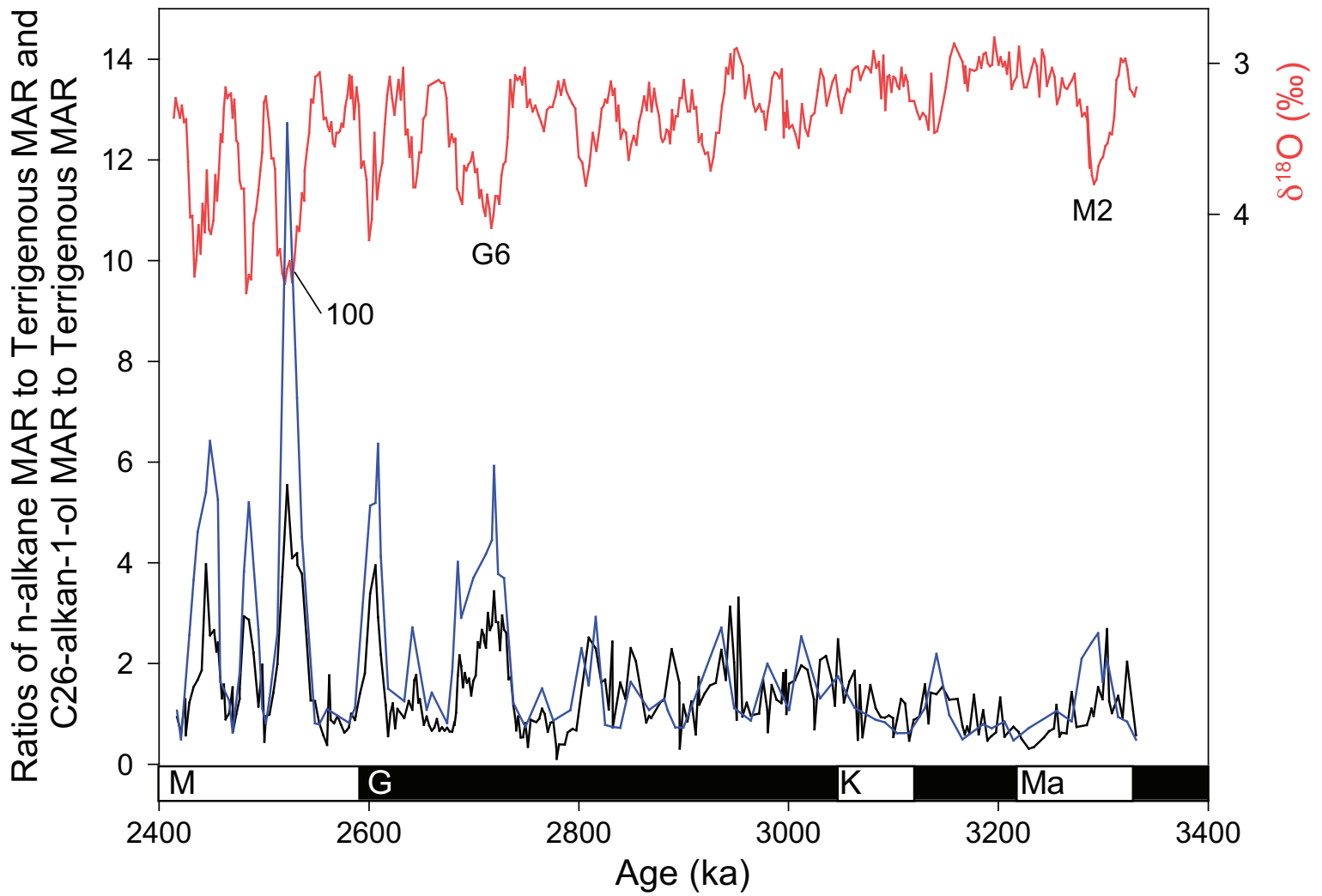
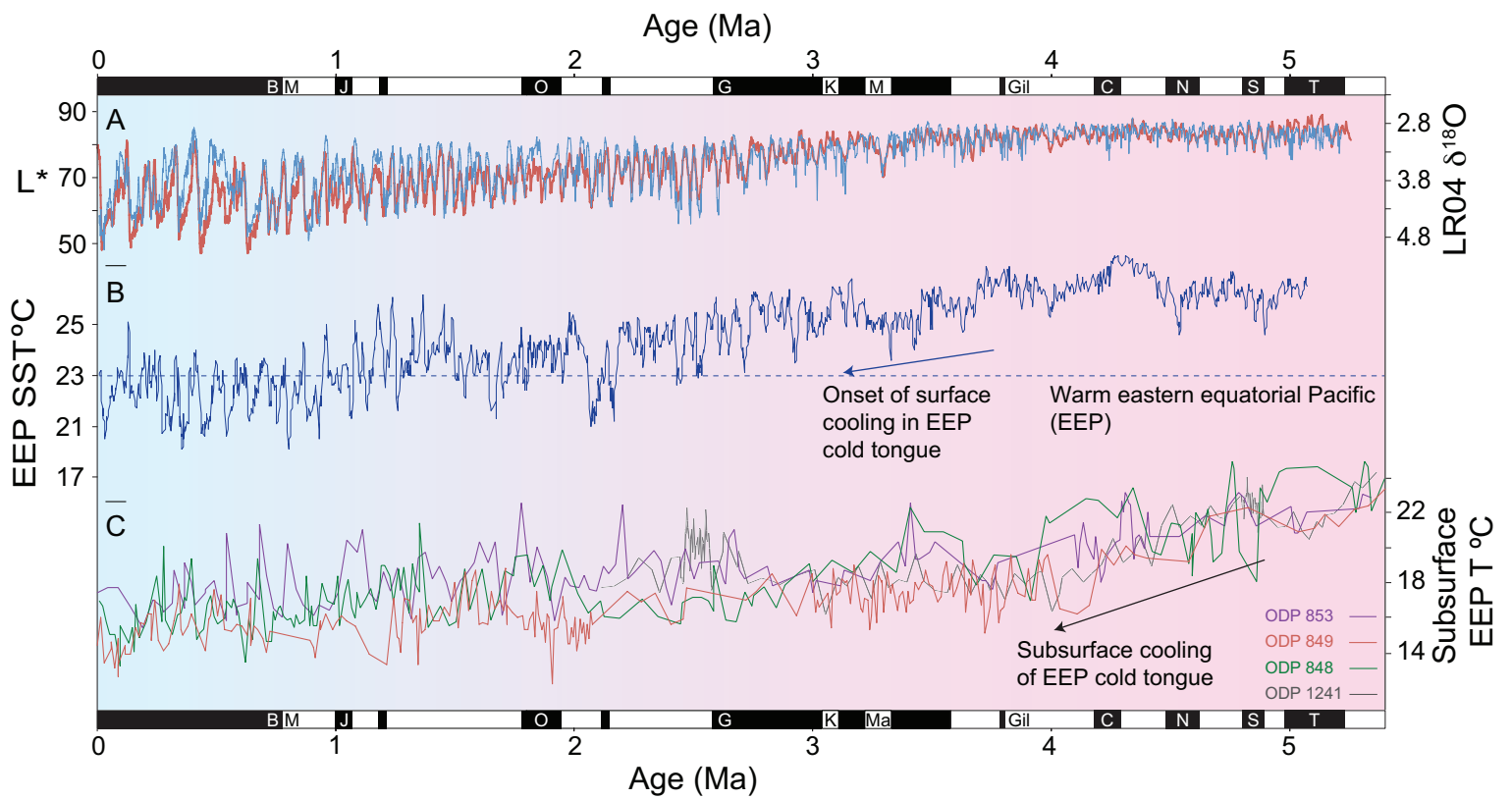


Figure 11



Supplementary Data

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'Equation File' for editorial office

[Click here to download Supplementary Data: Epsilon Nd equation.docx](#)