Aridification in continental Asia after the Middle Eocene Climatic Optimum (MECO)

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Global climate cooling from greenhouse to icehouse conditions occurred across an enigmatic transitional interval during the Eocene epoch characterized by incipient polar ice-sheet formation as well as short-lived warming events, of which the Middle Eocene Climatic Optimum (MECO) is most noticeable. Understanding this critical period requires high-resolution records that are being gathered in marine basins, but are still lacking in the terrestrial realm. Here, we provide a precisely-dated terrestrial record crossing the MECO time interval from the Xining Basin (NW China). We document a rapid aridification step and the onset of obliquity-dominated climate cyclicity indicated by lithofacies and pollen records dated at 40.0 Ma at the base of magnetochron C18n.2n. This shift is concomitant - within error - with the MECO peak warming in Ocean Drilling Program Site 1258 for which we reassessed the magnetostratigraphic age at 40.0 Ma (also at base of magnetochron C18n.2n). The rapidity of the shift observed in the Xining Basin and the region-wide aridification and monsoonal intensification reported around 40 Ma suggests Asian paleoenvironments were responding to global climate changes associated with the MECO. However, the Xining records show only the permanent shift but not the transient peak warming observed in marine MECO records. We thus relate this permanent aridification to occur during the post-MECO cooling. We propose the mechanisms linking global climate to Asian paleoenvironments may be eustatic fluctuations driving the stepwise retreat of the proto-Paratethys epicontinental sea or simply global cooling reducing moisture supply to the continental interior. In any case, Eocene global climate cooling from greenhouse to icehouse conditions seem to have played a primary role in shaping Asian paleoenvironments.

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1. Introduction

Ever since the first ocean sediments were drilled, hypotheses involving tectonic, oceanic and atmospheric dynamics have been proposed to explain the dramatic shift from greenhouse to icehouse conditions during the Cenozoic (DeConto and Pollard, 2003). Better comprehension is arising from compiled deep-sea temperature reconstructions from various ocean basins (Pälike et al., 2012) showing that after maximum temperatures in the early Eocene, ensuing long-term gradual cooling ultimately lead to major expansion of Antarctica ice-sheets at the Eocene–Oligocene Transition (EOT) 33.9 million years ago (Ma; Zachos et al., 2008). This transitional period of cooling is often referred to as the ‘doubthouse’ (Browning et al., 1996) and is characterized by short-lived warming events and by periodic polar ice-sheet formations. Recently, this period has received considerable attention because it is believed to represent the key interval to understand the long-term descent into the icehouse (Bohaty and Zachos, 2003; Pälike et al., 2012). The Middle Eocene Climatic Optimum (MECO) is the most prominent transient warming in this transitional period. The MECO was first identified in ocean drill cores from the Southern Ocean (Bohaty and Zachos, 2003). It has now been documented in stable isotope records worldwide, both in ocean drill sites in the Indian, Atlantic, Pacific and Southern Oceans and in land-based marine sections in Italy and the United Kingdom (Jovane et al., 2007; Villa et al., 2008; Luciani et al., 2010; Spofforth et al., 2010; Dawber et al., 2011). This clearly emphasizes the global impact of the MECO on the marine realm. The MECO is currently recognized as a ∼500-kyr warming event between ∼40.5 and ∼40 Ma characterized by gradual warming until sudden peak...
Regional paleoenvironmental context

The Xining Basin is a sub-basin in the western part of a broad Paleocene–Miocene basin complex along the north-eastern margin of the present-day Tibetan Plateau (Fig. 1a). After Mesozoic initial basin development, Cenozoic reactivation followed at ~50 and ~25 Ma due to the India–Asia collision (Dupont-Nivet et al., 2004; Horton et al., 2004). The basin provides a long and sub-complete stratigraphic record from ~50 to ~15 Ma, which has been well-dated using magnetostratigraphy (Dai et al., 2006; Xiao et al., 2010, 2012; Abels et al., 2011).

Previous sedimentary and pollen analyses from this basin have provided accurate records of stepwise regional aridification associated with global climate cooling culminating at the EOT (Dupont-Nivet et al., 2007, 2008; Xiao et al., 2010; Abels et al., 2011). Regular cyclicity of these sediments has been related to a dominant obliquity-forcing signal with additional impact of short eccentricity and interpreted as a sign for high-latitude climates controlling moisture supply to the area related to incipient polar ice-volume fluctuations (Xiao et al., 2010; Abels et al., 2011). In addition to the stepwise aridification of dry desert to steppe paleoenvironmental conditions, pollen analysis indicates long-term effect of regional uplift shown by the appearance of high-altitude Picea pollen in chron C16r at ~36.9 Ma (Dupont-Nivet et al., 2008; Hoorn et al., 2012). In consequence, aridification has been interpreted as a combination of changing moisture supply to the continental interior in response to global oceanic cooling, retreat and desiccation of the shallow epicontinental proto-Paratethys Sea to the west, as well as the long-term and stepwise regional uplift of various parts of the Tibetan Plateau (Dupont-Nivet et al., 2007; Abels et al., 2011; Bosboom et al., 2011, 2013).

3. Sections and sampling

Here, previously studied stratigraphic sections at Shuiwan and Xiejia (Dai et al., 2006; Dupont-Nivet et al., 2007; Abels et al., 2011) are extended downward and complemented with the new Tiefo and Dasigou sections (Figs. 1b and 2). Detailed lithological logs were made in the field. Samples were collected from evaporite beds for X-Ray Diffraction (XRD) mineralogy and from various lithologies throughout the sections for pollen analyses. XRD analyses and pollen extraction were performed using standardized techniques, which are described in detail in the Supplementary material.

Paleomagnetic sampling was performed using a standard portable electric drill powered by a portable gasoline generator. Samples were collected at an approximate resolution of 0.5 m and orientated with a magnetic compass mounted on an orientation stage. The field samples were cut into core specimens of approximately 10.5 cm³. Paleomagnetic analyses were carried out in the shielded Paleomagnetic Laboratory ‘Fort Hoofddijk’ of the Faculty of Geosciences at Utrecht University, the Netherlands. Representative specimens of characteristic lithologies were powderized and analyzed for their thermomagnetic behavior using a Curie balance (Mullender et al., 1993) and a KLY-3 susceptibility bridge. Single specimens were thermally demagnetized in a shielded oven by using up to 22 temperature steps of 5 to 50 °C (Supplementary Fig. S3). The remanent magnetization at each temperature step was measured by a 2G Enterprises DC SQUID cryogenic magnetometer.

4. Lithostratigraphy

The studied stratigraphic sections are characterized throughout the studied part of the basin by two distinct parts in terms of lithofacies, lithostratigraphic cyclicity and interpreted paleoenvironments (Figs. 2 and 3). The upper part consists of regular algal–lacustrine successions of the Xining Basin in NW China reveal significant paleoenvironmental change, while magnetostratigraphic dating ensures accurate age calibration and comparison with marine records.
Abels et al. (2011) has shown that the regular cyclicity is dominantly forced by obliquity (41-kyr) with additional influence of short eccentricity (100-kyr).

In the lower part, dry mudflat and saline lake lithofacies are also dominant, but lithologies are much more variegated. The regular cyclicity is absent and strong lateral variations preclude basin-wide bed-to-bed correlations. In particular, the prominent evaporitic interval comprising laminated gypsum, anhydrite, glauberite beds and thin interbedded clay layers is recognized as a perennial saline lake system with variable facies and age throughout the basin (Fig. 3e; see Fig. 2 for XRD sample levels and Supplementary Fig. S1 for XRD bulk results). At the Dasigou section, this interval includes a several-meter thick massive sandstone bed indicative of point-bar deposition in a fluvial channel. Also exclusively occurring in the lower part are decimetre-scale silt- and sandstone beds typical of ephemeral fluvial deposits, centimetre-scale layers of calcium-carbonate interpreted as chemical precipitates in a lacustrine environment and a hydromorphic paleosoil horizon showing color mottling, rootlets and bioturbation (Fig. 3c).

The transition from the lower to the upper part is characterized by mudstone-gypsum alternations, which can only be partially correlated across the basin due to lateral variation in depositional environments. The start of the transitional interval is at cycle −23, while the first bed that can be straightforwardly correlated between the four studied sections in the basin is cycle −15, marking the beginning of the upper part of the studied stratigraphic interval (Figs. 2, 3a and 3d).

Lithofacies in the lower part clearly indicate wetter atmospheric conditions during which a more hydrodynamic depositional environment led to higher morphological variability in the basin. Conversely, in the upper part, drier conditions are in line with the depositional environment becoming significantly more stable with individual sedimentary cycles correlatable across the studied part of the basin.

Following previous interpretations of younger regional lithofacies steps recorded in the Xining Basin (Dupont-Nivet et al., 2007; Abels et al., 2011), we attribute this change to regional atmospheric aridification. Furthermore, the observed aridification step is combined with the installation of obliquity-driven sedimentary cyclicity expressed up to the EOT, which have been previously attributed to high-latitude signal that could be enhanced by the occurrence of incipient polar ice sheet formation at this time (Xiao et al., 2010; Abels et al., 2011).

5. Palynology

To further ground our paleoenvironmental interpretation, we compiled a composite palynological record from different lithological beds throughout the Shuiwan, Tiefo and Xiejia sections (Fig. 5; Supplementary Tables S1 to S3 and Fig. S2). Nearest living relatives are indicated where possible and taxa were grouped according to biome type following Yu et al. (2000) and Ni et al. (2010). Four main groups are distinguished labeled Steppe–Desert, Angiosperms–Gymnosperms, Broad-leaved Forest types, and Ferns. Gymnosperms are very rare and the Angiosperms–Gymnosperms group thus nearly consists of angiosperms only (see Supplementary Table 1 for the composition of these groups).

The lower part of the succession is characterized by an average 50% presence of the Steppe–Desert group, around 20–25%
Angiosperms–Gymnosperms, 20–25% Broad-leaved Forest types, and on average 5% Ferns. Particularly taxa such as Ulmipollenites, but also Sapindaceae, and to a lesser extent the Retitricolpiites group, are abundant in these levels. The nearest living relative of Ulmipollenites is Ulmus (elm), a genus that presently occurs in the Asian steppe-desert and is common in fluvial beds or where ground water is nearby (Dulamsuren et al., 2009; Wesche et al., 2011). Ulmipollenites (included in the Broad-leaved Forest group) is very abundant at the top of the prominent evaporite interval in the Tiefo section. At the base lies the prominent white saline lake evaporite unit. e. Radial glaebrite crystals in a green mudstone matrix in the pre-MECO evaporite unit at the base of the Tiefo section (hammer for scale).

6. Magnetostratigraphic dating

To precisely date the identified lithofacies and pollen change in the Xining record, we performed magnetostratigraphy on four parallel sections that directly underlie and overlap the Shuiwan and Tashan sections previously dated from ∼39 Ma up well into the Oligocene ∼26 Ma (Abels et al., 2011; Xiao et al., 2012).

6.1. Rock magnetism

To determine the rock magnetic properties and carriers of the collected samples and to devise the best procedure for subsequent thermal demagnetization, rock magnetic analyses were performed first. From these analyses two clearly distinct behavioral types can be distinguished (Supplementary Fig. S3). Most of the samples from dry mudflat deposits (red mudstones) generally show a decrease in magnetization near the Curie temperature of magnetite (580°C), maghemite (~640°C) and infrequently of hematite (680°C), in agreement with previous studies (Dai et al., 2006; Xiao et al., 2010). Saline lake deposits (comprising muddy evaporite beds and greyish-green mudstones) were also sampled. They generally show a decrease in magnetization at temperatures considerably below or close to the Curie temperature of magnetite (580°C). In some samples, an increase in magnetization around 500°C is interpreted to relate to the oxidation of iron-sulphides into magnetite (Passier et al., 2001).

6.2. Thermal demagnetization

Single specimen samples from the sections were thermally demagnetized to reveal the remanent magnetization. For the preferably sampled red mudstones, after progressive removal of an overprint component at temperatures of 100 to 300°C, the trajectories of the characteristic remanent magnetization (ChRM) show linear decay towards the origin from 300°C to around 600°C and infrequently around 700°C, supporting the combined presence of magnetite and hematite as dominant ferromagnetic carriers in agreement with the rock magnetic results. Samples from evaporites also have an overprint up to 300°C usually followed by decay toward the origin around 500°C, occasionally until 600°C. This high temperature decay is often noisy because of the typically weak ChRM intensities of these samples and the apparent magnetic mineral transformation. Therefore, samples with a high temperature reversed polarity were clearly identified, but samples showing a high temperature normal polarity component were sometimes ambiguous and rejected from further analysis if the secondary normal overprint was not clearly separated from the higher temperature component.

6.3. ChRM direction analyses

For most samples, the ChRM directions were calculated by line-fits on orthogonal demagnetization diagrams using principal component analysis (Kirschvink, 1980) on a minimum of four temperature steps. Maximum angular deviations (MAD) on the line-fits were usually below 15°, but MAD of up to 30° were accepted if the polarity could be clearly discerned. For a few weaker reversed samples, typically from evaporite beds, the line-fits were forced through the origin if directions clustered in a distinct reversed position, and great circle ChRM analysis (McFadden and McElhinney, 1988) was performed if the demagnetization path was non-linear but along a great circle.

Virtual geomagnetic poles (VGMs) calculated from the ChRM directions show two distinct positive latitude (normal) and negative latitude (reversed) groups. VGMs outlying more than 45° from the mean normal and reversed VGP respectively were neglected for determining polarity zones (open points in Fig. 2). Using Fisher statistics (Fisher, 1953) the means of remaining normal and reversed ChRM directions were calculated (Supplementary Fig. S4). The Dasigou dataset passes the reversals test of McFadden and McElhinney (1990) with classification C. The reversals test is negative for Tiefo and Xiejia sections as the angle between the means of normal and reversed polarity is larger than the critical angle. The offset is 4.4° for Tiefo and 24.8° for Xiejia and interpreted to
be related to a partial overlapping normal overprint, expressed especially in the weaker ChRM gypsiferous sediments of the saline lake type. This overprint, however, does not affect the reliability of the magnetostratigraphy as previously noted for Xining Basin records (Dupont-Nivet et al., 2007, 2008; Abels et al., 2011).

6.4. Correlation to GPTS

The polarity zones were defined based on at least two consecutive VGP latitudes of identical polarity such that isolated directions within an interval of opposing polarity are neglected (Fig. 2). Apart from the lower Xiejia section consisting mainly of evaporites, the remarkably correlative polarity zonation between the Tiefo, Dasigou and upper Xiejia sections (Fig. 5) attests for the primary origin of the paleomagnetic signal and the reliability of the following straightforward correlation to the Geologic Time Scale 2012 (GTS12; Gradstein et al., 2012) from ∼44 Ma to ∼38 Ma (chrons C20 to C17).

The obtained pattern is the direct downward continuation of the previously correlated magnetostratigraphies of the Shuiwan and Tashan sections (Abels et al., 2011; Xiao et al., 2012) in
Fig. 5. Atmospheric aridification in the Xining Basin after the MECO event. Correlation of the four lithological sections in the Xining Basin by magnetostratigraphic correlations to the GTS12. To the right, the composite pollen record is displayed against time, showing permanent aridification is coincident with the lithological aridification step. To the left, stable isotope stratigraphy and magnetostratigraphy from ODP Site 1051 (Edgar et al., 2010) are given, confirming the approximate coincidence of the MECO peak warming with the aridification step on the Asian continent. See Fig. 2 for the legends of litho- and magnetostratigraphic symbols.

Younger than which a reliable magnetostratigraphic correlation is determined from C8n.2n (26 Ma) down to C18n.2n (39 Ma). Accordingly, the upper two normal intervals are correlated to chron C18n and C17n, like in the lithologically correlated lateral Shuiwan section. The long reversed interval below at Tiefo and Dasigou likely represent the expected C18r and C19r. The expected occurrence of the short C19n is represented by a few normal samples consistently occurring at correulative levels of the Tiefo and Dasigou sections. At the bases of Dasigou and Tiefo sections, the subsequent long normal chron represents the next normal C20n underlain by C20r at the Tiefo section (Fig. 2; Supplementary Fig. S5).

In the newly developed temporal framework, the environmental shift and onset of cyclicity can be dated at 40.05 ± 0.08 Ma assuming constant accumulation rates through C18r up to the position of the lithofacies shift of cycle −15 and given the uncertainties in the positions of the top and base of C18r respectively. Alternatively, a very similar age of 40.00 ± 0.08 Ma is obtained for the shift if the top and base of C18n (rather than C18r) are used as a reference for the calculation. This supports the robustness of our age determination, although the real uncertainty range is likely to be larger due to the assumption of constant sedimentation rates in the Xining Basin over the duration of magnetochron C18r or C18n. Therefore, we place the absolute age for the onset of the cyclicity...
(occurring in the base of chron C18n.2n) at 40.0 ± 0.1 Ma using GTS12. Similarly, the start of the preceding transitional interval at cycle −23, in the top of chron C18r, is dated at 40.41 ± 0.11 Ma or 40.41 ± 0.15 Ma for C18r and C18n, respectively, which leads us to use an age of 40.4 ± 0.2 Ma for further discussion (Fig. 5).

To compare our terrestrial record with global climate records, we reassess the age of the MECO event in Ocean Drilling Program (ODP) Site 1051, which is the only unambiguous record of the MECO directly coupled to high-resolution magnetostratigraphy (Fig. 4; Bohaty et al., 2009; Edgar et al., 2010). As uncertainty intervals were not given in the original study, we re-interpreted the paleomagnetic data from this site by calculating the ChRM directions and VGPs from the majority of the original raw demagnetization data, similar to our own paleomagnetic analyses of the Xining Basin sections. After cut-off of VGPs outlying more than 45° from the mean, we obtain a polarity pattern very similar to that of Edgar et al. (2010), but now including uncertainty intervals. As the top of C18n.2n is lacking from the data, we calculated the position of the MECO with respect to the base of C18r by again assuming constant accumulation rates through C18r up to the position of the MECO and given the uncertainties in the positions of the top and base of C18r respectively. Our magnetostratigraphic calibration gives a position within chron C18n.2n of 113.0 ± 3.6% with respect to the base of chron C18r, yielding an age of the MECO peak warming at 40.01 ± 0.04 Ma assuming constant accumulation rates through the well-defined magnetochron C18r below. Direct comparison of this age with obtained ages for the environmental shift in the Xining record (40.0 ± 0.1 Ma) suggests that the Asian aridification occurred roughly concomitant with the peak warming of the MECO event at 40.01 ± 0.04 Ma (Fig. 5). However, a probably real uncertainty of 0.2–0.4 million years, hampered by the assumption of constant sediment accumulation rates over the long chron C18r, are likely on these ages, preventing further determination whether the Asian aridification occurred directly before, during, or after MECO peak warming.

7. Discussion

A permanent aridification step dated approximately concomitant in time with the transient peak warming of the MECO in the marine realm (Bohaty et al., 2009) is evident in our Asian terrestrial sedimentary records. However, a transient rather than a permanent change would be expected in the Xining Basin records, because the MECO has been associated to a peak in pCO2 (Bijl et al., 2010) that should affect continental paleoenvironments. This may indicate that thresholds to changing depositional environments in the Xining Basin were not reached during the MECO event, suggesting change in atmospheric climate during the MECO was not very severe. However, the absence of a transient record may be attributed to the gap in our pollen record across the MECO event and/or attributed to a depositional hiatus during the MECO transient peak although accumulations are relatively constant throughout the interval from chron C20n (41 Ma) up to C8n.2n (26 Ma) in this part of the Xining Basin.

The Xining Basin records have revealed further aridification steps similar to the one observed at the MECO. The most prominent occurs in the top of C17n.1n at ~37.0 Ma where lithofacies and pollen indicate significant aridification (Abels et al., 2011; Hoorn et al., 2012), which has shown to be concomitant with high-altitude pollen appearance (Dupont-Nivet et al., 2008). The subsequent larger shift occurs at the onset of the Eocene–Oligocene greenhouse to icehouse transition at ~33.9 Ma (Dupont-Nivet et al., 2007; Xiao et al., 2010) with gypsum bed disappearance interpreted as atmospheric aridification. These aridification steps occurred within an depositional environment dominated by dry mudflat and saline lakes experiencing changes from warmer and wetter environments to colder and especially more arid conditions in the Xining Basin. However, the aridification step at the MECO (~40.0 Ma; base of chron C18n.2n) presented here marks the onset of these dry mudflat and saline lacustrine paleoenvironments replacing the underlying more hydrodynamic sedimentary environments with wetter facies and palynology. This suggests this step is a prominent event, which is probably regionally extensive as explained below.

Indeed, this well-dated record of climate deterioration in the Xining Basin is regionally corroborated by other Asian sedimentary records with less age accuracy. Song et al. (2013) interpret climate deterioration in the neighboring Qaidam Basin, with clear long-term trends from warm and wet climate in the middle Eocene to drier environments in the Oligocene. Here a drop of thermophilic palynological taxa (defined by Lu et al., 2010, as consisting of ferns like Pterisissipores, Polypondiacæaesporites, gymnosperms such as Podocarpidites, Tsugaepollenites, and Rutaceopollis) and decrease in chemical weathering around the base of chron C18n.2n at 40 ± 1 Ma are interpreted as indicative of significant aridification. Similar to the Xining Basin, this major aridification step is reported to occur after a transient warmer and wetter interval that can be tentatively correlated to the transient peak warming of the MECO and the aridification to the post-MECO cooling. In another section from the Qaidam Basin, aridification starting also at around the base of chron C18n.2n at ~40 Ma is indicated by heavier stable carbon isotopes of lake carbonate (Rieser et al., 2009). From Fushun, north-eastern China, Quan et al. (2011) report palynological data indicating East Asian monsoon intensification with an age attributed at 41–40 Ma after the late middle Eocene. The authors link the intensification to uplift of parts of the Tibetan Plateau functioning as a rain shadow prohibiting eastern moisture to penetrate further west into the Asian continent.

However, the onset of Asian desertification has been associated with monsoon intensification caused by the complex interplay between Tibetan Plateau uplift, global cooling and westward retreat of the shallow epicontinental proto-Paratethys sea (Ramstein et al., 1997; Guo et al., 2002; Dupont-Nivet et al., 2007, 2008; Bosboom et al., 2011; Huber and Goldner, 2011). The stepwise nature and the rapidity of the changes observed in the Xining Basin favors a swift mechanism such as global climate and/or epicontinental sea retreat rather than the gradual tectonic uplift of the Tibetan Plateau, although these probably worked in concert upon reaching threshold conditions (Dupont-Nivet et al., 2008; Hoorn et al., 2012). Oceanic global cooling may have had a direct influence on aridification by reducing moisture supply to the continental interior. More indirectly, successive sea-level lowering induced by ice-sheet formation linked to global cooling may have governed the stepwise retreat of the shallow epicontinental sea from Central Asia (Ramstein et al., 1997; Dupont-Nivet et al., 2007; Zhang et al., 2007). Recent study shows that the stepwise sea retreat to the west and the stepwise aridification recorded in the Xining Basin are closely correlated in time (Bosboom et al., 2011, 2013), which suggests that the retreat has been an important forcing mechanism driving the desertification of Asia and may provide the mechanism linking global climate cooling to Asian aridification.

We therefore speculate that the observed shift in the Asian record is linked to the relatively rapid post-MECO cooling in the marine realm (Iyle et al., 2005; Villa et al., 2008; Bohaty et al., 2009; Pallik et al., 2012), rather than to the transient warming during the MECO event, although both are within age uncertainty range. A link with global cooling is supported by the observed appearance of obliquity cycles after the aridification step in the Xining record. This requires a climatic mechanism dominated by obliquity without significant influence of precession. However, at these lower latitudes, the direct influence of obliquity on solar insolation is small with respect to the influence of pre-
8. Conclusions

Our results provide the first precisely-dated terrestrial record across the transitional late Eocene interval preceding the Eocene–Oligocene Transition in continental China. We find in lithofacies and pollen of the Xining Basin the expression of a sudden permanent aridification with a shift to obliquity-dominated climate cyclicity at the base of chron C18n.2n around 40 Ma. This prominent aridification step appears to be regionally correlative to an Asian paleoenvironmental transition associated with monsoonal intensification. We suggest that the stepwise nature of the Asian paleoenvironmental changes indicates they are driven by global climate changes in concert with regional tectonic uplift, epicontinental sea retreat, and monsoonal intensity change. A link to global cooling is supported by the appearance of obliquity-dominated cycles most likely related to high-latitude climate variability. This may be speculatively related to large ice volume variability during incipient polar ice-sheet formation reported to start at this time. Rather than the transient MECO warming event, the observed aridification step could thus better be linked to post-MECO cooling marking the onset of the final descent into the icehouse, ultimately leading to the Eocene–Oligocene greenhouse to icehouse transition. To ascertain these speculations, additional high-resolution records leading to the Eocene–Oligocene greenhouse–icehouse transition are required.

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Appendix A. Supplementary material

Supplementary material related to this article can be found online at http://dx.doi.org/10.1016/j.epsl.2013.12.014.

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