

# A 9 myr cycle in Cenozoic $\delta$ 13C record and long-term orbital eccentricity modulation: Is there a link?

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4	A ~9 myr cycle in Cenozoic $\delta^{13}$ C record and long-term orbital eccentricity
5	modulation: Is there a link?
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24	Abstract
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26	The ~65-myr-long Cenozoic carbon isotope record ( $\delta^{13}$ C) of Zachos et al. (2001
27	2008) documents a strong long-term cycle with a mean pseudoperiodicity close to ~9 my

This cyclicity modulates the ~2.4 myr eccentricity cycle amplitude, hinting at a possible link between long-term astronomical and geological variations. Some phase shifts between ~9-myr  $\delta^{13}$ C and astronomical cycles suggest that additional processes (e.g., tectonics) contribute to these long-term carbon-cycle variations.

The strong response of  $\delta^{13}$ C to long-term eccentricity periods (~9 myr, ~2.4 myr, ~400 kyr) supports the hypothesis that the long time-residence of carbon in the oceans amplifies lower frequency or dampens higher frequency orbital variations. Additionally, the strong expression of low-amplitude ~9 myr eccentricity cycle in the  $\delta^{13}$ C record could be explained by energy-transfer process from higher to lower frequency cycles, and all eccentricity components modulate the carrier climatic precession cycles.

Finally, the Paleocene-Eocene Thermal Maximum (PETM, 55.9 Ma) event, which corresponds to a pronounced  $\delta^{13}$ C negative excursion, is situated within a strong decrease in the most prominent ~9 myr  $\delta^{13}$ C cycle, hinting at a link between accelerated rates in  $\delta^{13}$ C variations and the PETM. This specific ~9 myr  $\delta^{13}$ C cycle seems to be amplified by non-orbital mechanisms in atmosphere-continent-ocean system, such as previously suggested methane release from gas hydrate and volcanism.

**Keywords:** Cenozoic deep-sea  $\delta^{13}$ C records, multi-millionyear cyclicity, ~9 myr cyclicity, astronomical driving force, eccentricity.

#### 1. Introduction

It has been widely demonstrated that Earth's orbital parameters (climatic precession, obliquity, eccentricity) control paleoclimate changes induced by variations in solar radiation intensity received by our planet (Hays et al., 1976; Imbrie et al., 1984). Research has documented the Earth's orbital parameters in the geologic record over broad time scales, from millennial to million year time scales. Today, there is a broad consensus that insolation forcing from the Earth's orbital parameters is a major external driver of changes in atmospheric, continental, and oceanic conditions across all latitudes. Accordingly, recent research has now turned to explain extreme short-lived climate events (e.g., Lourens et al., 2005), ice-sheet formation and carbon reservoir cycling (e.g., Pälike et al., 2006a), biotic extinctions (van Dam et al., 2006), and other poorly understood phenomena. Reciprocally, geophysical processes, such as a post-glacial rebound (Laskar et al., 1993; Mitrovica et al., 1997) and mantle convection (Forte & Mitrovica, 1997), could have influenced secular variations in the dynamic ellipticity of the Earth, which consequently influence the astronomical precession constant value (e.g., Berger et al., 1992).

In this study, we focus on very long-term periodic variations in the carbon isotope record ( $\delta^{13}$ C) of Cenozoic deep-sea sediments by which we suggest a possible link with long-term astronomical modulation cycle. Numerous Cenozoic and Mesozoic studies have interpreted these long-term carbon cycle variations as trends, most of them suggesting a tectonic origin (e.g., Zachos et al., 2001; Leckie et al., 2002; Li et al., 2009). Through a comparison between long-term  $\delta^{13}$ C cycle and orbital eccentricity modulation cycle, we suggest another alternative, i.e., orbital driving force.

Additionally, our results concur with and build on previous studies pointing to the long residence-time of carbon in oceans amplified by longer orbital periods (Pälike et al., 2006a). At the same time, we do not rule out possible interactions between tectono-oceanic and climatic variations (e.g., Raymo and Ruddiman, 1992; Lagabrielle et al., 2009) in carbon recycling, triggered or retroacted by the astronomical driving force.

## 2. Cenozoic carbon isotope record ( $\delta^{13}$ C)

Building on ocean drilling during the last three decades, ~65-myr-long Cenozoic carbon and oxygen records ( $\delta^{13}$ C and  $\delta^{18}$ O) were compiled from benthic foraminifera isotope data from more than 40 deep-sea cores of the Deep Sea Drilling Project (DSDP) and Ocean Drilling Program (ODP) (Zachos et al., 2001, 2008, Fig. S1, and Cramer et al., 2009). Today, these long records serve as a basic reference for numerous Cenozoic paleoclimate studies that address rhythms, trends and events in Earth's changing climate (Zachos et al., 2001). The compiled records of Zachos et al.'s (2008) compilation exhibit variable resolution, which is comprised between less than ~1 kyr and ~8 kyr for the last ~34 Ma, and could reach 40 kyr for the last 35 to 65 Ma interval (Zachos et al., 2008). This resolution of the stacked data is however sufficient for time-series analysis of low-frequency (multi-millionyear) variations. In this study, we examine the behavior of long-period cycles in the  $\delta^{13}$ C record (Fig. 1), and in particular, we suggest a possible link with long-term eccentricity cycling throughout the Cenozoic (Section 3).

The ~65-myr-long Cenozoic carbon record shows evidence of ~9-myr cycles that we denote as Cb1 to Cb8 (Fig. 1). Zachos et al.'s (2001) studies were focused on oxygen isotope evolution to interpret temperature and glacial trends and short-lived events such as the Paleocene-Eocene Thermal Maximum (PETM), Oi-1, Mi-1 glaciation events, etc. The Oi-1 and Mi-1 events delimit the Cb4 cycle, which roughly spans the Oligocene Epoch. This cycle was studied in detail by Pälike et al. (2006a) at a ~4-kyr sampling resolution (Fig. 2a), who demonstrated the strong expression of the ~400-kyr cycles in the carbon isotope signal. Filtering and modulation analysis studies were focused mainly on the ~100-kyr eccentricity and 41-kyr obliquity bands to recover the important ~1.2 and ~2.4-myr astronomical

modulations. Modulation behavior of the lower frequency ~400-kyr and ~2.4-myr eccentricity cycles (Section 3.2) were not examined.

In fact, because the Oligocene interval is only 11.9-myr long (from 22 to 33.9 Ma), Pälike et al. (2006a) did not consider Cb4 to be a true cycle (Section 3). Instead, they subdivided the interval into 4 phases, which they interpreted as long-term trends, through comparison with the oxygen isotope record (see their Figure 1). Here, we show that this cycle, Cb4, contains four ~2.4-myr cycles easily correlatable to their analogs in astronomical solutions (Laskar et al., 2004, and Laskar et al., 2011a). The ~2.4-myr term acts to modulate the ~400-kyr cycles, while the 'Cb4' modulates the 2.4-myr cycles (Figs. 1 and 2a, Section 3.2).

#### 3. Methods and results

#### 3.1. Spectral analysis

## 3.1.1. Spectral analysis of $\delta^{13}$ C data

Spectral analysis of the raw  $\delta^{13}$ C record of Zachos et al.'s (2008) compilation shows two dominant peaks centered on periods of 27 and 8.4 myr (Fig. 3a). Similar results are obtained when we consider the same interval (i.e., Cenozoic) in Cramer et al.'s (2009)  $\delta^{13}$ C compilation (Fig. 3b). The 8.4 myr cyclicity is prominent in the  $\delta^{13}$ C records and represent the cycles denoted Cb1 to Cb8 (Fig. 1). Its pseudo-period could vary from ~7.5 myr (Cb2) to ~11 myr (Cb3). However, the 27 myr peak is not evident in  $\delta^{13}$ C time series, but it may correspond to the two cycles delimited by the minima of Cb1, Cb4 and Cb7 (Figs. S1 and S2). These very long-term  $\delta^{13}$ C oscillations have a significant effect on spectral results. When we subtract them, other peaks at higher frequency band are strengthened, but still with low

power because of the high-power ~9 myr peak (Fig. S2). To highlight low- and highfrequency bands, we display the raw  $\delta^{13}$ C spectrum, associated with the robust noise, on a logarithmic scale (Figs. 3c,d). Zachos et al.'s (2008) compilation (Fig. 3c) shows several spectral peaks around the 400 kyr band, but none of them matches the astronomically predicted and well defined 405 kyr eccentricity term; one peak centered on the period of 1.2 myr, likely related to long-term obliquity modulation cycle; and two other peaks at 1.9 and 2.5 myr periods, may represent the 2.4 myr orbital eccentricity term. Cramer et al.'s (2009) compilation (Fig. 3d) shows also several peaks around the 400 kyr band, only one peak (404 kyr, see also Fig. S2) may correspond to the 405 kyr eccentricity period. This mismatch between  $\delta^{13}$ C and astronomical periodicities may be due to the sampling rates in these stacked  $\delta^{13}\text{C}$  data (see Section 3.2). Differences in spectral analysis results between the two δ<sup>13</sup>C compilations (i.e., Zachos et al., 2008; Cramer et al., 2009) concur with this hypothesis of the sampling-rate effect. Nevertheless, the multi-myr  $\delta^{13}$ C cycles are robust and persistent in both compilations (Figs. 3a,b). Particularly, the ~9 myr cycles are well detected by lowpass and bandpass filtering of the two  $\delta^{13}$ C time series (Fig. 1). The 405 kyr and 2.4 myr  $\delta^{13}$ C cyclicities are documented with confidence in the single ODP site 1218 (Fig. 2a, Pälike et al., 2006a), which respectively reflect 'g2-g5' and 'g4-g3' eccentricity terms (where g2, g3, g4, and g5 are respectively related to the precession of the perihelions of Venus, the Earth, Mars, and Jupiter, Fig. 4). However, neither 8.4 nor 27-myr cycles is evident in the Earth's orbital variations (e.g., Laskar et al., 2004). To determine if these long-term cycles have an astronomical origin, we tested their presence in two astronomical models, La2004 and the most recent and constrained La2010d (Laskar et al., 2004; Laskar et al., 2011a).

#### 3.1.2. Spectral analysis of the orbital eccentricity variations

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The objective of this section is to test if there are small (low amplitudes) low-frequency (periods longer than ~2.4 myr) cyclicities in the raw eccentricity time series. Thus, we applied spectral analysis (Figs. 4a,b) to the La2004 and La2010d orbital eccentricity for the last 67 Ma (time interval equivalent to that in  $\delta^{13}$ C records).

The La2004 model was demonstrated as precise for the last 40 Ma (Laskar et al., 2004). The La2010d is the most constrained version of the La2010 astronomical model that extends the precision to the last 50 Ma (Laskar et al., 2011a,b). The main difference between La2004 and La2010 models is that the La2004 initial conditions are ajusted to the JPL numerical ephemeris DE406 (Standish, 1998) over –5000 yr to +1000 yr from the present date, while La2010 is fitted to the most precise planetary ephemeris INPOP06 and INPOP08 (Fienga et al., 2008, 2009). In particular, La2010d version is adjusted to the INPOP06 over the last 1 Ma that includes the five major asteroids, Ceres, Vesta, Pallas, Iris, and Bamberga. La2010a, La2010b and La2010c are fitted to the INPOP08 over only 580 kyr for La2010a and La2010b, and over 1 myr for La2010c. However, the latter does not include the five major asteroids, Ceres, Vesta, Pallas, Iris, and Bamberga. Certain orbits of these asteroids underwent a strong chaotic behavior, and consequently their motion will be unpredictable over 400 kyr, which in turn influences the Earth's orbital eccentricity calculation (for detail see Laskar et al., 2010b). Thus, the La2010d version is considered as the most constrained version in the La2010 model (Laskar et al., 2010b).

Frequency decomposition in harmonic analysis of both La2004 and La2010d raw eccentricity are very similar (Figs. 4a,b). However, the relative amplitudes of spectral peaks are slight different; this difference is linked to the parameters used in each astronomical model (detailed in Laskar et al., 2011a). At the low frequencies, amplitude spectrum (Fig. 4b) shows peaks around the 400-kyr band, i.e., (g2 - g5) and  $(g2 - g5) \pm (g4 - g3)$  terms

composed of the three periods 405 kyr for (g2-g5), 489 kyr for (g2-g5)-(g4-g3), and 345 kyr for (g2-g5)+(g4-g3) (Fig. 4, see Laskar et al., 2004, their table 6). The interference of the two coupled periods (405 with 345 kyr, and 405 with 489 kyr) induces the long ~2.4 myr eccentricity term, i.e., (g4-g3), which is represented by a significant peak (Fig. 4b, see Section 3.2.1). Two other main peaks at 670 and 959 kyr correspond respectively to (g2-g1) and (g1-g5) terms. Finally, a weak peak centered on the period of ~4.5 myr corresponds to another eccentricity term discussed in Section 3.2.1.

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Lowpass filtering does not show significant low-frequency (longer than ~4.5 myr) variations given that, if present, they are with very low energy as illustrated in the amplitude spectra (Fig. 4b). Nevertheless, low-amplitude astronomical variations could have significant influence on Earth's climate change. As an example, though the direct effect of eccentricity forcing on insolation is relatively low, the strong imprint of the main eccentricity cycles (the average 97, 128 kyr, and the strong single 405 kyr; Fig. 4a) in paleoclimatic variations could be indirect, i.e., via energy transfer from the carrier climatic precession into the modulator eccentricity. Same behavior could manifest in the very low-frequency eccentricity terms (Section 3.2.1), where energy could be transferred from higher to lower frequency terms via amplitude/frequency modulation, and all eccentricty terms modulate, in turn, the precession index cycles. The most compelling example is that the  $\sim$ 2.4 myr eccentricity term (q4 - q3) with relatively low amplitude (e.g., Laskar et al., 2004) is well recognized in geological records (Olsen and Kent, 1999; Hilgen et al., 2003; van Dam et al., 2006; Mitchell et al., 2008; Abels et al., 2010; and many others), while it could act as a modulator of higher frequency eccentricity terms, either at the 100 k.y. or at the 400 k.y. cycle bands (e.g., 405 with 489 kyr, and 405 with 345 kyr, Fig. 4b, Section 3.2.1). Another striking example concerns the Earth's obliquity variations. In particular, the ~1.2 myr obliquity cycle is strongly documented in Cenozoic icehouse strata (e.g., Wade and Pälike, 2004; Pälike et al., 2006a), although it only acts as a modulator of the higher frequency obliquity cycles (i.e., p + s3 ≈ 41

kyr and p + s4  $\approx$  39.6 kyr). Thus, these examples indicate that the presence of a long-period astronomical cycle with low amplitude, but with a strong imprint in a paleoclimatic record is possible via amplitude/frequency modulation of the higher frequency cycles, and the consequent energy-transfer process. Therefore, we tested possible very long-period eccentricity cycles via amplitude modulation (AM) technique.

We performed AM analysis on both the raw  $\delta^{13}C$  data and the orbital eccentricity, in order to seek possible analogs. We focused on the ~400-kyr and ~2.4-myr cycles because of their strength in the  $\delta^{13}C$  record (Fig. 2a), even if they seem to be biaised by the stacked sampling rates (Figs. 3c,d), and low resolution of the early Cenozoic  $\delta^{13}C$  data (Fig. S3).

#### 3.2. Amplitude modulation (AM) analysis

First, we bandpass-filtered the orbital eccentricity ~100-kyr, ~400-kyr and ~2.4-myr variations, and performed Hilbert transforms (e.g., p. 435 in Hinnov, 2000) on the filtered series (Fig. 1). Then, we applied the same procedure to the  $\delta^{13}$ C record, but only at the longer periods, i.e. ~400-kyr and ~2.4-myr (Fig. S3), because the  $\delta^{13}$ C data resolution is too low to adequately recover ~100-kyr scale behavior through the past 65 Ma, and the stacked sampling rates have a significant effect on higher frequency band (Figs. 3c,d; Section 3.1). Finally, we applied spectral analysis on the amplitude modulation (AM) envelopes (Figs. 5 and 6).

## 3.2.1. AM of the orbital eccentricity

Time-series analysis of astronomical AM envelopes shows significant long-term modulations in the eccentricity bands. The AM envelope of the short eccentricity (~100-kyr) shows strong 0.405- and 2.4-myr modulations (Fig. 5a), whereas those of the ~400-kyr

eccentricity shows strong ~2.4-myr and lesser ~4.5-myr modulations (Figs. 5b and S4). The AM envelope of the ~2.4-myr orbital eccentricity band shows ~9-myr and ~26-myr cyclicity (Figs. 5c,d). The latter is not obvious in the AM time series. However, the former is well detected in AM time series, and shows some correlation with the  $\delta^{13}$ C record (Section 4). Thus, the ~4.5- and ~9-myr cyclicities are present in the orbital eccentricity modulations. The ~2.4-myr component could derive from the interference of the two coupled close frequencies in the 400 kyr band (Fig. 4b). In particular, the interferences of 405 kyr (g2 – g5) with 345 kyr ((g2 – g5) + (g4 – g3)), and 405 kyr (g2 – g5) with 489 kyr ((g2 – g5) – (g4 – g3)) induce both the 2.4 myr term (g4 – g3) (see Fig. 4b). The astronomical origin of the ~4.5-myr cycles was discussed in Laskar (1990) as deriving from the resonant argument  $\theta = 2(g_4 - g_3) - (s_4 - s_3)$ , where  $g_3$ ,  $g_4$  are related to the precession of the perihelions of the Earth and Mars,  $s_3$ ,  $s_4$  are related to the precession of the same planets. The ~4.5-myr cyclicity is present in the orbital eccentricity solution (Fig. S5). However, the ~9-myr was not previously explicited in the astronomical models.

## 3.2.2. AM of the $\delta^{13}$ C variations

Then, we performed ~400-kyr and ~2.4-myr AM analysis on the  $\delta^{13}$ C records of Zachos et al.'s (2008) and Cramer et al.'s (2009) compilations (Figs. 6 and S3). The spectrum of ~400-kyr AM of Zachos et al.'s compilation (Fig. 6a) shows a strong significant peak centered on 5 myr, and two other weaker but significant peaks of periods 3.8 and 1.9 myr. The 5- and 3.8-myr peaks may represent the average of the ~4.5-myr periodicity (discussed previously). The 1.9 myr peak could represent a distorted ~2.4-myr eccentricity modulation term (see below). The spectrum of ~400-kyr AM of Cramer et al.'s compilation (Fig. 6b) indicates two strong significant peaks centered on 2.4 and 16 myr. The former

corresponds likely to 'g4-g3' eccentricity term (Fig. 5b). However, the latter is not evident neither in the astronomical variations nor in the  $\delta^{13}$ C time series.

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The spectrum of ~2.4-myr AM of Zachos et al.'s compilation (Fig. 6c) shows mainly four significant peaks (above 99% CL) centered on the periods of 6.3, 8.1, 12, and 32 myr. The three first ones, with an average of 8.8 myr, may represent the ~9 myr astronomical variations. The latter again is not evident in the  $\delta^{13}$ C time series. The spectrum of ~2.4-myr AM of Cramer et al.'s compilation (Fig. 6d) highlights three significant peaks centered on the periods of 7.4, 10.2, and 27.5 myr. The former two, with an average of 8.8 myr, most likely represent the ~9 myr astronomical variations. The latter may represent the peak seen in the raw  $\delta^{13}$ C spectrum (i.e., 29 myr, Fig. 3b). Therefore, AM results suggest that the ~9-myr cyclicity modulates the ~2.4-myr and not the ~400-kyr cycles as has been demonstrated in the astronomical models. Additionally, AM results of the 2.4 myr band is more consistent than those of the 400 kyr band. This again suggests the more effect of sampling rates on higher frequency cycles. In sum, despite of the Cramer et al.'s (2009) compilation show some  $\delta^{13}$ C frequencies, in the ~2.4-myr AM, consistent with astronomical frequencies, the large differences of AM outputs (Figs. 6 and S3) between the two  $\delta^{13}$ C versions (Zachos et al., 2008; Cramer et al., 2009) preclude any positive conclusion regarding AM results. The stacked sampling rates could be the most important factor that affects the documentation of higher frequency cycles, and hence AM use, given that both  $\delta^{13}$ C compilations are rescaled to the same timescale (GTS2004, Gradstein et al., 2004). Nevertheless, the ~9 myr cycles are prominent and robust in both  $\delta^{13}$ C compilations.

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## 4. $\delta^{13}$ C variations and orbital eccentricity correlation

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The 9-myr eccentricity cycles correlate well with the raw  $\delta^{13}$ C (Fig. 1) because of the higher sensitivity of global carbon-cycle to eccentricity forcing (e.g., Cramer et al., 2003; Pälike et al., 2006a; Abels et al., 2010). A clear negative correlation between eccentricity and  $\delta^{13}$ C cycles was observed from ~8 to ~60 Ma. Maxima of  $\delta^{13}$ C correspond to minima of ~9 myr eccentricity cycles. Similar negative correlation was also demonstrated at the higher frequency eccentricity cycles (~2.4-myr, ~400-kyr, and ~100-kyr, Cramer et al., 2003; Pälike et al., 2006a, and many others). Cb1 to Cb8  $\delta^{13}$ C cycles correlate well with the ~9 myr astronomical cycles (Fig. 7). The average cross phase estimate between ~2.4-myr eccentricity AM and ~9 myr  $\delta^{13}$ C cycles is about -180 degrees (Fig. 7). This supports the antiphase relationship between eccentricity and  $\delta^{13}$ C variations. However, visual examination of the time series (Fig. 1, Cuves 3 and 4) indicates that  $\delta^{13}$ C and eccentricity variations are sometimes phase-shifted at the ~9 myr cyclicity suggesting that non-orbital processes (e.g., tectonics) could interfere into these long-term  $\delta^{13}$ C oscillations (e.g., Zachos et al., 2001; Leckie et al., 2002; Li et al., 2009). On the other hand, La2004 astronomical model (Laskar et al., 2004) is valid for only the last 40 Ma, while the La2010d model (Laskar et al., 2011a) is reliable for the last 50 Ma. Thus, correlation between  $\delta^{13}$ C and astronomical variations for the interval 50 to 67 Ma (Cb7 and Cb8) is critical, and should be considered with caution till a future extended astronomical model will be available, and future studies could enhance geological timescale resolution.

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Besides, we note a positive correlation between  $\delta^{13}$ C and astronomy for the last ~8 Ma. This exception may be due to the compilation of the carbon isotopes data itself, which should be checked in future  $\delta^{13}$ C compilations. In fact, possible unreliable  $\delta^{13}$ C compiled data could exist for the Plio-Pleistocene where basin to basin fractionation develops (low values of the Pacific, J. Zachos, *pers. comm*). Recent studies concur with this hypothesis, and show that Atlantic-Pacific  $\delta^{13}$ C differential responses exist for the last ~10 Ma (Cramer et al., 2009,

their figure 6). A different hypothesis, Wang et al. (2010) suggested that the expression of eccentricity in the  $\delta^{13}$ C record is obscured for at least the last ~1.6 Ma, related to a major change in the oceanic carbon reservoir associated with a restructuring of the Southern Ocean. Finally, Katz et al. (2005) interpreted a long-term linear trend in this interval (precisely for the last ~17 Ma, in their figure 1) as the result of a closing phase of the Wilson cycle, when continents reassemble and the Atlantic Ocean basin closes. According to these observations, we restrict our interpretation, in terms of possible orbital forcing of the 9-myr  $\delta^{13}$ C cycles, to the interval ~8 to ~50 Ma, and with caution the interval ~50 to ~60 Ma, where 9-myr  $\delta^{13}$ C and eccentricity cycles are anticorrelated.

#### 5. Discussion: how does orbital forcing drive Cenozoic carbon cyclicity?

High-amplitude (1‰) eccentricity-scale cyclicity has been described in various upper Paleocene-Neogene  $\delta^{13}$ C records (Woodruff and Savin, 1991; Flower and Kennett, 1995; Diester-Haass, 1996; Zachos et al., 1996, 1997, 2001; Cramer et al., 2003; Pälike et al., 2006a,b), suggesting that carbon recycling through the atmosphere-biosphere-ocean reservoir (e.g., Siegenthaler and Sarmiento, 1993; Sarmiento and Bender, 1994) preferentially responds to orbital eccentricity from modulation of the climatic precession. For example, by applying climatic precession forcing to carbonate/organic carbon fluxes and deposition, and the residence-time of carbon in the atmosphere-biosphere-ocean, Cramer et al. (2003) modelled a theoretical eccentricity curve consistent with interpreted eccentricity cycles (~100 and ~400 kyr) in their measured bulk carbonate  $\delta^{13}$ C. Specifically, they showed a correspondence between maxima in ~400-kyr eccentricity and transient decreases in the bulk carbonate  $\delta^{13}$ C during the late Paleocene - early Eocene. In another example from the Oligocene, Pälike et al. (2006a) demonstrated that long-term insolation forcing could modulate atmospheric  $CO_2$  levels as well as the oceanic carbonate ion ( $CO_3^2$ ) concentration,

leading to expansion and contraction of biospheric productivity in tune with long-term orbital periodicities. Specifically, Pälike et al. (2006a) highlighted a strong ~400-kyr signal in the  $\delta^{13}$ C record, which they explained as the consequence of the long residence-time of carbon in the oceans, leading to enhanced sensitivity to longer forcing periods.

In this study, we show a prominent ~9 myr cyclicity in  $\delta^{13}$ C record, which may correspond to ~9 myr AM cycles in orbital eccentricity variations. This is in accordance with the idea of long residence-time of carbon in amplifying longer orbital periodicities. We also note that the maxima of long-term  $\delta^{13}$ C oscillations correspond to minima of long-term eccentricity modulations (Figs. 1 and 2a), which again concurs with previous  $\delta^{13}$ C cyclostratigraphic studies (Cramer et al., 2003; Pälike et al., 2006a, Rickaby et al., 2007; and many others). Therefore, if there is a link between  $\delta^{13}$ C record and orbital eccentricity modulation at this multi-millionyear timescale, and considering previous studies in carboncycle modeling, we could suggest that eccentricity induced insolation through its modulation of the climatic precession may play an important role in carbon cycling, controlling a chain of astro-climatically sensitive processes such as terrestrial weathering, biosphere productivity, carbonate sedimentation and dissolution, burial and oxidation of organic carbon, etc (e.g., Westbroek et al., 1993; Cramer et al., 2003; Kurtz et al., 2003; Pälike et al., 2006a; Rickaby et al., 2007; Li et al., 2009). At the same time, observed phase shifts between ~9-myr eccentricity and  $\delta^{13}$ C cycles suggest that additional processes (e.g., tectonics) could contribute to the  $\delta^{13}$ C variations.

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Finally, the most prominent ~9-myr  $\delta^{13}$ C cycle (Cb7, Figs. 1 and 2a), which corresponds to a well expressed ~9-myr astronomical cycle in the eccentricity modulation, includes the Paleocene-Eocene Thermal Maximum (PETM) event (Figs. 1 and 2b). In particular, the PETM, which corresponds to a pronounced  $\delta^{13}$ C negative excursion (2.5-3‰), is situated within a strong decrease in the Cb7  $\delta^{13}$ C cycle, hinting at a possible link between

 $\delta^{13}$ C rates and the occurrence of the event. The Cb7 cycle, especially its decreasing part, seems to be amplified compared to the other ~9 myr  $\delta^{13}$ C cycles (Fig. 1). This observation is beyond the scope of our study, and needs, however, future work to model causes of rates in the  $\delta^{13}$ C variations and their link with the PETM onset in this specific interval of Cenozoic (e.g., volcanism: Storey et al., 2007, and J. Zachos, pers. com; methane release from gas hydrate: Dickens et al., 1995, 1997).

#### 6. Conclusions

We show that Cenozoic carbon isotope record ( $\delta^{13}$ C) exhibits a multi-millionyear cycle of a pseudo-periodicity close to 9 myr. Amplitude modulation (AM) analysis of astronomical models shows that the ~2.4 myr eccentricity cycles are mainly modulated by ~9 myr cycles similar to those observed in the  $\delta^{13}$ C record. Several lines of correlation are observed between ~9 myr eccentricity envelopes and  $\delta^{13}$ C cycles pointing to a possible link between the two.

Additionally, the Paleocene-Eocene Thermal Maximum (PETM) event, which corresponds to a pronounced  $\delta^{13}C$  negative excursion, coincides with a critical interval in the ~9 myr  $\delta^{13}C$  cycle. An unusual strong decrease in the most prominent ~9 myr  $\delta^{13}C$  of the Cenozoic may reflect specific processes in atmosphere-contient-ocean system (e.g., methane release from gas hydrate, volcanism), responsible for the amplification of the ~9 myr  $\delta^{13}C$  cycle and the PETM excursion.

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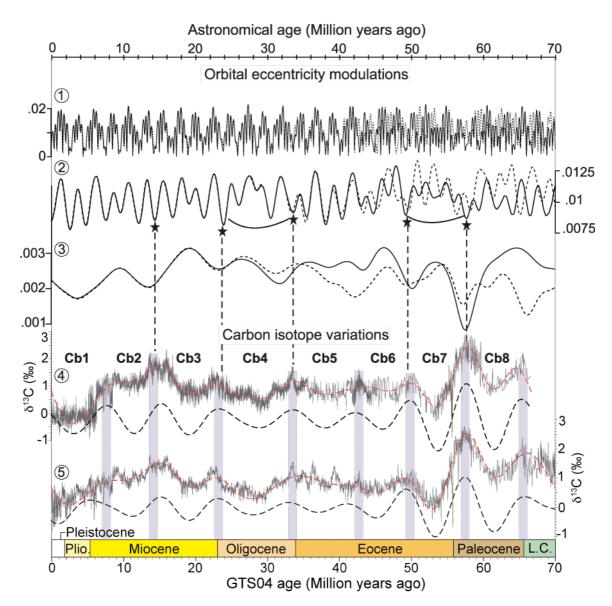


Figure 1: Amplitude modulations (AM) of the nominal La2004 (solid curves) and La2010d (dashed cuves) astronomical models (Laskar et al., 2004, and Laskar et al., 2011a), and correlation with the ~9 myr δ<sup>13</sup>C cyclicity of deep-sea δ<sup>13</sup>C records (Zachos et al., 2001, 2008; Cramer et al., 2009). Top to bottom, curve 1: AM of the short eccentricity band (9.4 ±2 myr<sup>-1</sup>), curve 2: inverted AM of the ~400-kyr eccentricity band (2.5 ±0.5 myr<sup>-1</sup>), curve 3: AM of the ~2.4-myr eccentricity band (0.4245 ± 0.1350 myr<sup>-1</sup>). curves 4: raw δ<sup>13</sup>C record (Zachos et al., 2001, 2008), also shown low-pass (0 to 0.17 myr<sup>-1</sup> band) and bandpass (0.1265 ±0.04245 myr<sup>-1</sup>) filtered δ<sup>13</sup>C record, the ~9-myr δ<sup>13</sup>C oscillations are shown (labelled Cb1 to Cb8 delimited by vertical bars), curves 5: δ<sup>13</sup>C record (Cramer et al., 2009) with 5 point running average. Low-pass (0 to 0.17 myr<sup>-1</sup> band) and bandpass

 $(0.1265 \pm 0.04245 \text{ myr}^{-1})$  filtered  $\delta^{13}\text{C}$  record is also shown, applied to the raw  $\delta^{13}\text{C}$  data (resampled to 0.004 myr). For filtering, we used a Gaussian filter (Paillard et al., 1996). All filtering in orbital eccentricity was carried out on the -10 to 75 Ma interval to avoid filter-edge effect. AM analysis was performed by Hilbert transformation (e.g., p. 435 in Hinnov, 2000). The vertical dashed lines represent the correlation between the 'Cb' cycles and the astronomical ~9-myr cycles, with emphasis on five tie-points (represented with black stars) which show good correlation of Cb3, Cb4, and Cb7 to their equivalent astronomical cycles (vertical dashed lines: possible bundling of four ~2.4 myr cycles).

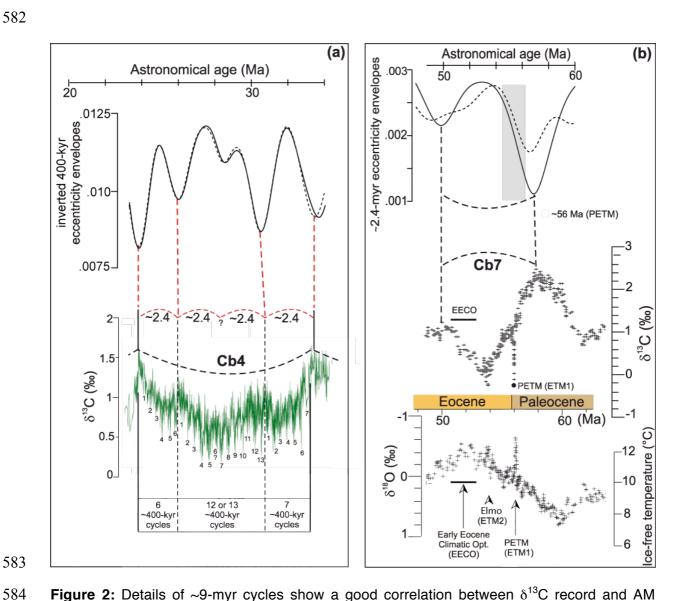


Figure 2: Details of ~9-myr cycles show a good correlation between  $\delta^{13}C$  record and AM eccentricity, and the position of  $\delta^{13}C$  brief events ( $\delta^{18}O$  is also shown, from

Supplementary Figure S1). (a) The Cb4  $\delta^{13}$ C cycle correlates to the ~9-myr astronomical cycle (from Fig. 1, curve 2). The two middle ~2.4-myr cycles are "amalgamated" in both the  $\delta^{13}$ C and the astronomical signal. This supports the validity of the astronomical models. (b) The Cb7  $\delta^{13}$ C cycle correlates to the ~9-myr astronomical cycle (from Fig. 1, curve 3), but should be considered with caution given that neither the geological time scale nor the astronomical models are precise in this time interval. The vertical bar indicates the interval (~54.5 to ~56.2 Ma) including the PETM event (56 Ma, estimates are from Westerhold et al., 2007, 2008, 2009, and Kuiper et al., 2008; 55.9 Ma according to Charles et al., in press). Both the PETM and the Elmo (ETM2) events are situated in the decreasing part of the strongest ~9-myr  $\delta^{13}$ C cycle (Cb7) with the PETM is near the inflection-point (~56 Ma).

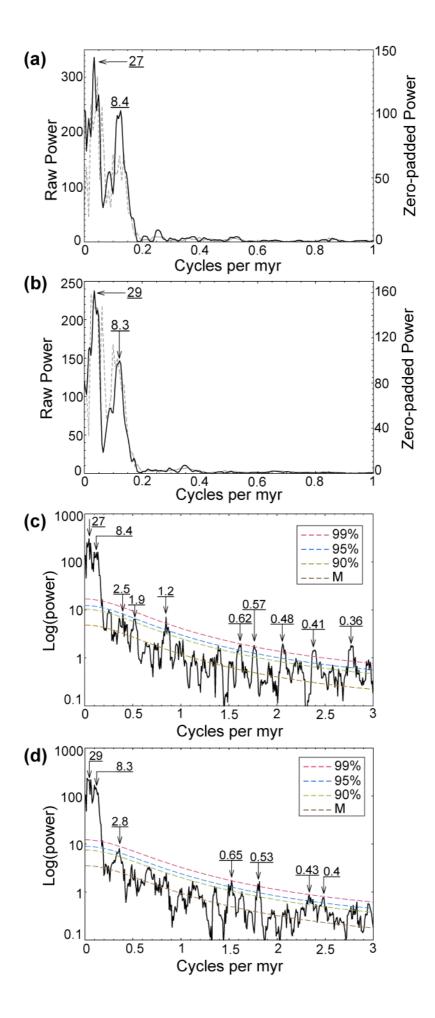


Figure 3: 2π-MTM power spectra of the δ¹³C records (Zachos et al., 2008; Cramer et al., 2009), using the multitaper method (Thomson, 1982) as implemented in SSA-MTM Toolkit (Ghil et al., 2002). Results of noise modelling (also shown) were estimated using linear fitting and median filtering over 20% of the average Nyquist frequency range. (a) Zachos et al.'s (2008) compilation, dashed line: raw δ¹³C record linearly interpolated to a ~4-kyr sample rate (the average sampling interval of the record is ~4.7 kyr), solid line: 1x zero-padded δ¹³C used to highlight the ~8.4-myr peak (see also Fig. S2). (b) Cramer et al.'s (2009) compilation, dashed line: raw δ¹³C record linearly interpolated to a ~4-kyr sample rate (the average sampling interval of the record is ~4.7 kyr), solid line: 1x zero-padded δ¹³C used to highlight the ~8.3-myr peak (see also Fig. S2). (c) Spectrum of (a) in a logarithmic format over a broader frequency band, together with the modeled red noise spectrum. (d) Spectrum of (b) in a logarithmic format over a broader frequency band, together with the modeled red noise spectrum.

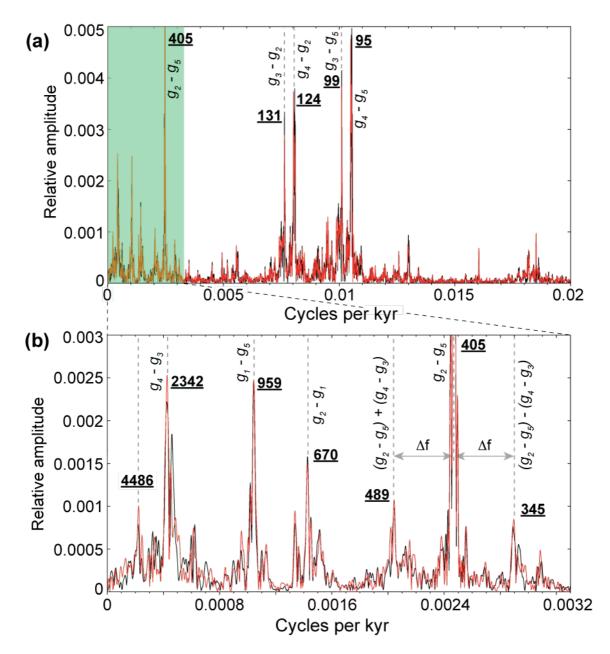


Figure 4: 2π-MTM harmonic analysis of the orbital eccentricity in the La2004 (black line) and La2010d (red line) astronomical models (Laskar et al., 2004, 2011a) over the last 67 Ma (time interval equivalent to that in δ¹³C data). (a) the raw spectra displayed over frequencies 0 to 0.02 cycles/kyr, and truncated amplitude axis at 0.005: only the peak representing the 405 kyr eccentricity term is truncated (its full relative amplitude is ~0.010). He truncated power axis to highlight peaks at low frequencies in the shaded area. (b) a zoom of the shaded area in 'a' to well visualize low-frequency peaks, the amplitude axis is also truncated at 0.003. All peaks are labelled in kyr, and the origin of each eccentricity component is also shown, where g1, g2, g3, g4 and g5 are the secular

frequencies related respectively to the precession of the perihelions of Mercury, Venus, the Earth, Mars, and Jupiter (see Laskar et al., 2004 for detail). Note that only the main peaks depicting the main eccentricity components are shown.  $\Delta f$  in 'b' equals to 0.000427 cycles/kyr, which corresponds to a period of 2342 kyr, i.e., (g4-g3) is the result of two possible interferences of (g2-g5) with (g2-g5)-(g4-g3), and (g2-g5) with (g2-g5)+(g4-g3) (see text for discussion).



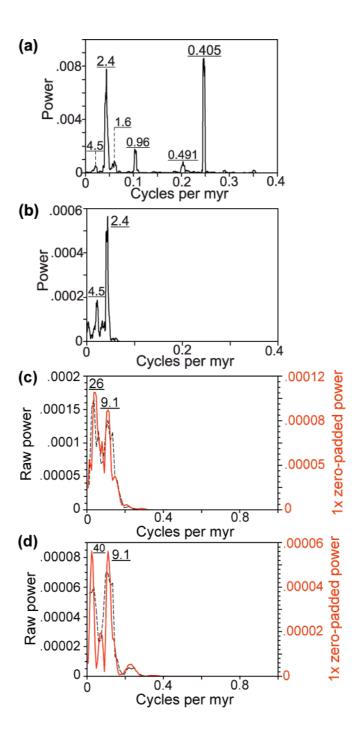


Figure 5: 2π-MTM power spectra of amplitude modulation (AM) of δ¹³C records (Zachos et al., 2008; Cramer et al., 2009), using the multitaper method (Thomson, 1982) as implemented in SSA-MTM Toolkit (Ghil et al., 2002). Results of noise modelling (also shown) were estimated using linear fitting and median filtering over 20% of the average Nyquist frequency range. (a) Spectrum of the AM envelopes of the ~400-kyr δ¹³C band of Zachos et al.'s (2008) compilation (Supplementary Figure S3, solid curve 5). (b) Spectrum of the AM envelopes of the ~400-kyr δ¹³C band of Cramer et al.'s (2009) compilation (Supplementary Figure S3, dashed curve 5). (c) Spectrum of the AM envelopes of the ~2.4-myr δ¹³C band of Zachos et al.'s (2008) compilation (Supplementary Figure S3, solid curve 4), dashed line: raw AM, solid line: 1x zero-padded AM. (d) Spectrum of the AM envelopes of the ~2.4-myr δ¹³C band of Cramer et al.'s (2009) compilation (Supplementary Figure S3, solid curve 4), dashed line: raw AM, solid line: raw AM, solid line: 1x zero-padded AM. All peaks are labelled in myr.

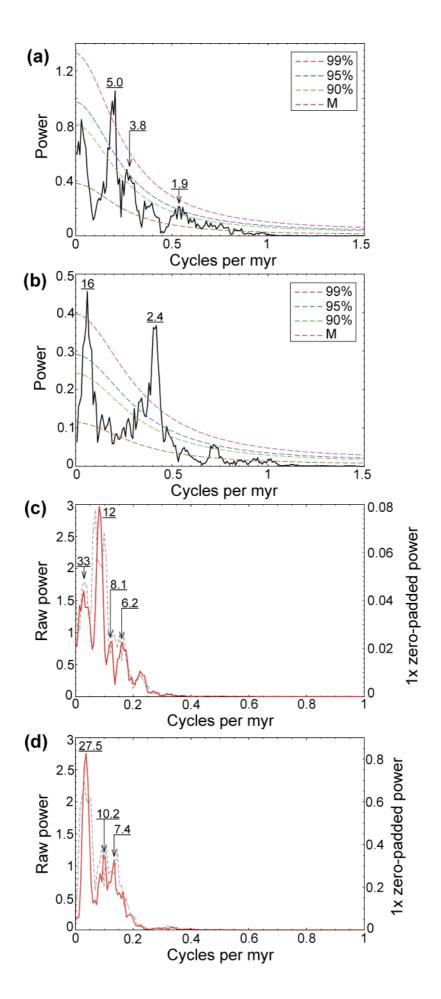


Figure 6: 2π-MTM power spectra of amplitude modulations (AM) of La2004 and La2010d orbital eccentricity (Laskar et al., 2004, 2011a), using the multitaper method (Thomson, 1982) as implemented in SSA-MTM Toolkit (Ghil et al., 2002). (a) AM of the ~100-kyr La2004 eccentricity band (Fig. 1, solid curve 1). (b) AM of the ~400-kyr La2004 eccentricity band (Fig. 1, solid curve 2). (c) Dashed line: AM of the ~2.4-myr La2004 eccentricity band (Fig. 1, solid curve 3), solid line: 1x zero-padded of ~2.4-myr La2004 eccentricity AM used to highlight the ~9-myr peak. (d) Dashed line: AM of the ~2.4-myr La2010d eccentricity band (Fig. 1, dashed curve 3), solid line: 1x zero-padded of ~2.4-myr La2010d eccentricity AM used to highlight the ~9-myr peak. All peaks are labelled in myr.

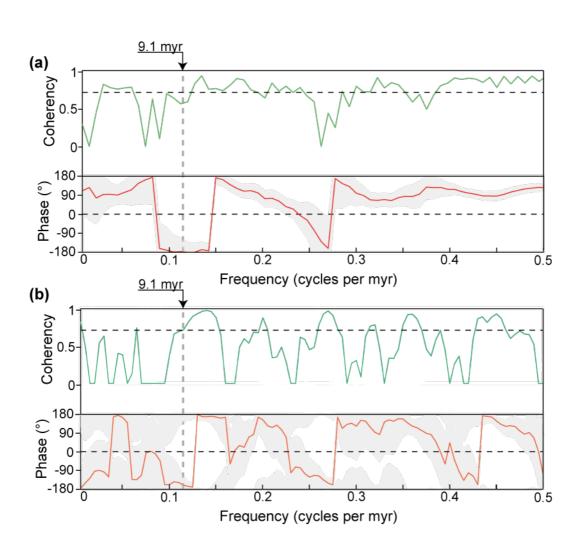


Figure 7: Coherency and cross-phase spectral analysis of envelopes of ~2.4 myr cycles of

eccentricity time series (Fig. 1, Curve 3) versus ~9 myr  $\delta^{13}$ C cycles (from the raw  $\delta^{13}$ C record of Zachos et al., 2008 in Fig. 1, Curve 4). (a)  $\delta^{13}$ C versus ~2.4 myr AM of La2004 model (Laskar et al., 2004). (b)  $\delta^{13}$ C versus ~2.4 myr AM of La2010d model (Laskar et al., 2011a). Note that  $\delta^{13}$ C and eccentricity are coherent at ~9 myr cyclicity, and that ~9 myr  $\delta^{13}$ C oscillations are ~180° out of phase with ~9 myr eccentricity cycles. The approximate 95% confidence level for the coherence between red noise and a narrow band signal is indicated by the dashed line; the zero phase line is indicated by the dashed line, and the approximate 95% confidence interval for phase is indicated by the gray shading. Vertical dashed lines indicate the ~9.1 myr frequency (see Figs. 5c,d). We used the cross-MTM method in the Matlab routine of Peter Huybers (e.g., Huybers and Denton, 2008).

## **Boulila et al.'s Supplementary Information**

## **Supplementary figures**

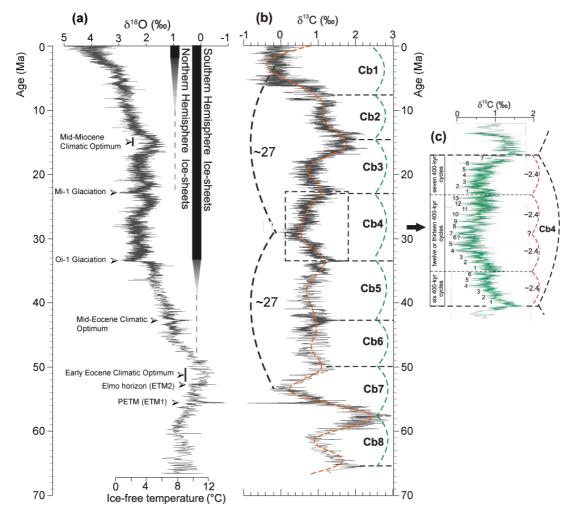


Figure S1: Global deep-sea oxygen and carbon isotope records based on data compiled from more than 40 Deep Sea Drilling Project and Ocean Drilling Program sites (Zachos et al., 2001, 2008). (a) Oxygen record, the main climatic events are also shown, ETM1 (also known as the Elmo horizon) and ETM2 (also known as PETM) represent Eocene Thermal Maximum events, with the PETM is the Paleocene-Eocene Thermal Maximum, the main hyperthermal event. Note that the δ<sup>18</sup>O temperature scale was computed for an ice-free ocean (~1.2 ‰ standard mean ocean water, SMOW). From the early Oligocene on, and possibly for some time before that, much of the variability in the δ<sup>18</sup>O values may reflect volume changes in polar ice sheets. The vertical bars provide a rough estimate of the ice volume in each hemisphere relative to the ice volume during the Last Glacial Maximum. (b) Carbon isotope record, pseudo-periodic ~9-myr (~7 to ~11 myr) δ<sup>13</sup>C cycles are shown (labelled Cb1 to Cb8). Possible ~27-myr δ<sup>13</sup>C cycles are also shown.

(c) A detailed Cb4 cycle, data from Pälike et al. (2006a) with ~4-kyr sampling intervals. We identified the ~400-kyr and ~2.4-myr cycles within Cb4. The two middle ~2.4-myr cycles are "amalgamated", as are the analogous astronomical ~2.4-myr cycles over this time interval (see Figs. 1 and 2a).

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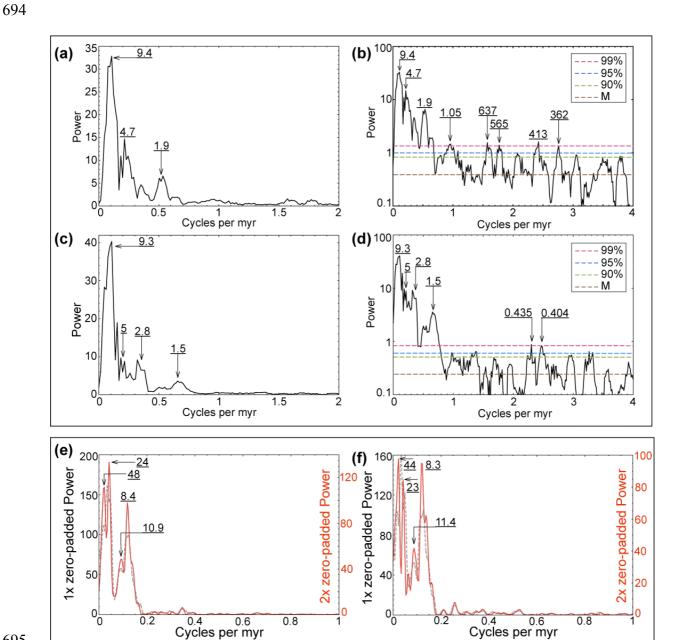


Figure S2: Upper panel: 2π-MTM power spectra of a selected interval (16.5 to 54.5 Ma) of Cenozoic  $\delta^{13}$ C records, after removal of a third-order polynomial fit to attenuate irregular long-term variations due to the ~27- and 29-myr oscillations (spectral peaks in main Fig. 3a,b). (a) Spectrum in a linear scale of the  $\delta^{13}$ C of Zachos et al.'s (2001, 2008)

compilation. (b) Spectrum of 'a' in a logarithmic scale over a broader frequency band, together with the modeled robust red noise spectrum. (c) Spectrum in a linear scale of the  $\delta^{13}$ C of Cramer et al.'s (2009) compilation. (d) Spectrum of 'c' in a logarithmic scale over a broader frequency band, together with the modeled robust red noise spectrum. Results of noise modelling: Curve M is the linear median-smoothed, fitted robust red noise spectrum; the upper 90%, 95% and 99% confidence limits. Note that the srongest spectral peak is centred on ~9 myr period in both compilations (Zachos et al., 2001, 2008, and Cramer et al., 2009). This supports the prominence and robustness of the 9 myr  $\delta^{13}$ C oscillations (see text for discussion). **Lower panel:**  $2\pi$ -MTM power spectra of Cenozoic  $\delta^{13}$ C records of Zachos et al.'s (2008) compilation (a) and Cramer et al.'s (2009) compilation (b). Spectra of 1x zero-padded time series (grey-dashed line as in main Figs. 3a,b) and spectra of 2x zero-padded time series (red solid line). Note that the 2x zero-padding enhances the resolution of the ~8.5 and ~25 myr peaks. The ~46 myr (48 myr in 'e' and 44 myr in 'f') peak may represent a single oscillation delimited by Cb2/Cb3 and Cb7/Cb8 boundaries (Fig. S1). We used the multitaper method (Thomson, 1982) as implemented in SSA-MTM Toolkit (Ghil et al., 2002). All peaks are labelled in myr.

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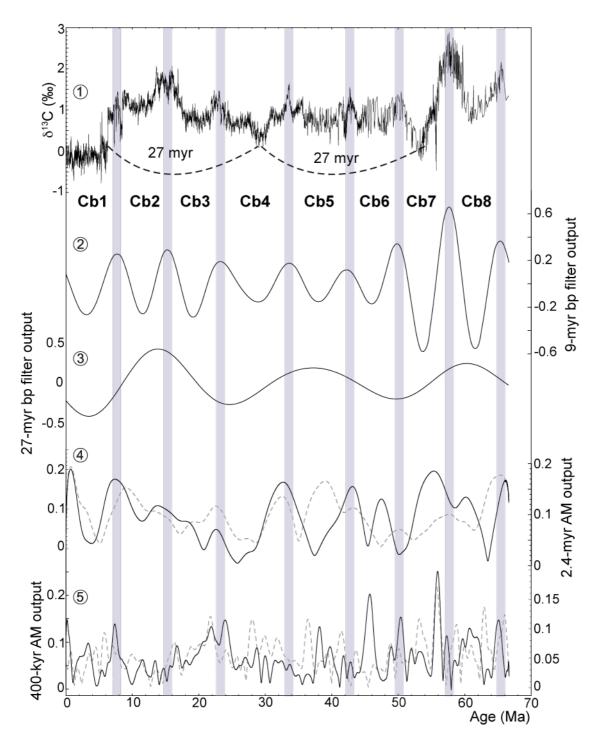
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**Figure S3:** Amplitude modulations (AM) time series of the raw  $\delta^{13}$ C records (Zachos et al., 2001, 2008, Cramer et al., 2009). From top to bottom: **curve 1**:  $\delta^{13}$ C record of Zachos et al. (2008) with 5-point running average, the ~9-myr  $\delta^{13}$ C pseudo-periodic oscillations (labelled Cb1 to Cb8) and the possible ~27-myr cycles are shown, **curve 2**: ~9-myr bandpass filter output of Zachos et al.'s (2008) raw  $\delta^{13}$ C (0.1265 ±0.0435 myr<sup>-1</sup> band), **curve 3**: ~25-myr bandpass filter output of Zachos et al.'s (2008) raw  $\delta^{13}$ C (0.048 ±0.015

myr<sup>-1</sup> band), **curve 4**: AM envelopes of the ~2.4-myr eccentricity band (0.4245  $\pm$  0.1350 myr<sup>-1</sup> band), solid black curve: AM applied to Zachos et al.'s (2008) raw  $\delta^{13}$ C, dashed-grey curve: AM applied to Cramer et al.'s (2009) raw  $\delta^{13}$ C, **curve 5**: AM envelopes of the ~400-kyr eccentricity band (2.5  $\pm$ 0.5 myr<sup>-1</sup> band), solid black curve: AM applied to Zachos et al.'s (2008) raw  $\delta^{13}$ C, dashed-grey curve: AM applied to Cramer et al.'s (2009) raw  $\delta^{13}$ C. All band-pass filtering was performed using a Gaussian filter (Paillard et al., 1996). Before analysis,  $\delta^{13}$ C data are resampled and linearly interpolated each 0.004 myr for both compilations.

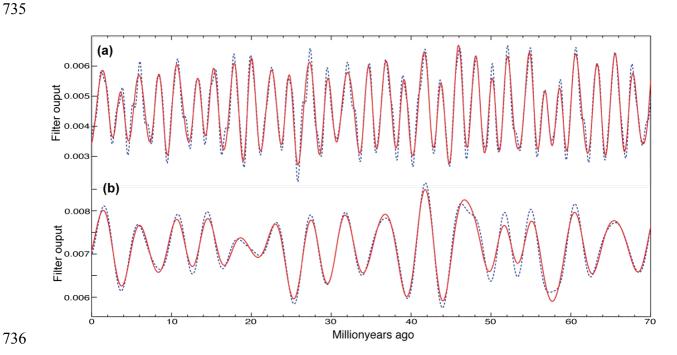


Figure S4: Low-pass filtering of the La2004 raw eccentricity time series (Laskar et al., 2004) to extract the resonant cyclicities corresponding to the argument  $\theta = 2(g_4 - g_3) - (s_4 - s_3)$  (Laskar, 1990; Laskar et al., 1992), where  $g_3$ ,  $g_4$  are related to the precession of the perihelions of the Earth and Mars,  $s_3$ ,  $s_4$  are related to the precession of the nodes of the same planets. (a) ~2.4 myr eccentricity cycles extracted with two different frequency-cutoff bands, a moderate band (0 to 0.7 cycles/myr, solid line) and a large band (0 to 0.5 cycles/myr, dashed line). (b) ~4.5 myr eccentricity cycles extracted with two different

frequency-cutoff bands, a moderate band (0 to 0.28 cycles/myr, solid line) and a large band (0 to 0.33 cycles/myr, dashed line). All low-pass filtering was performed using the Taner filter (Taner, 2000).