

Supplementary Material

A record of seafloor methane seepage across the last 150 million years

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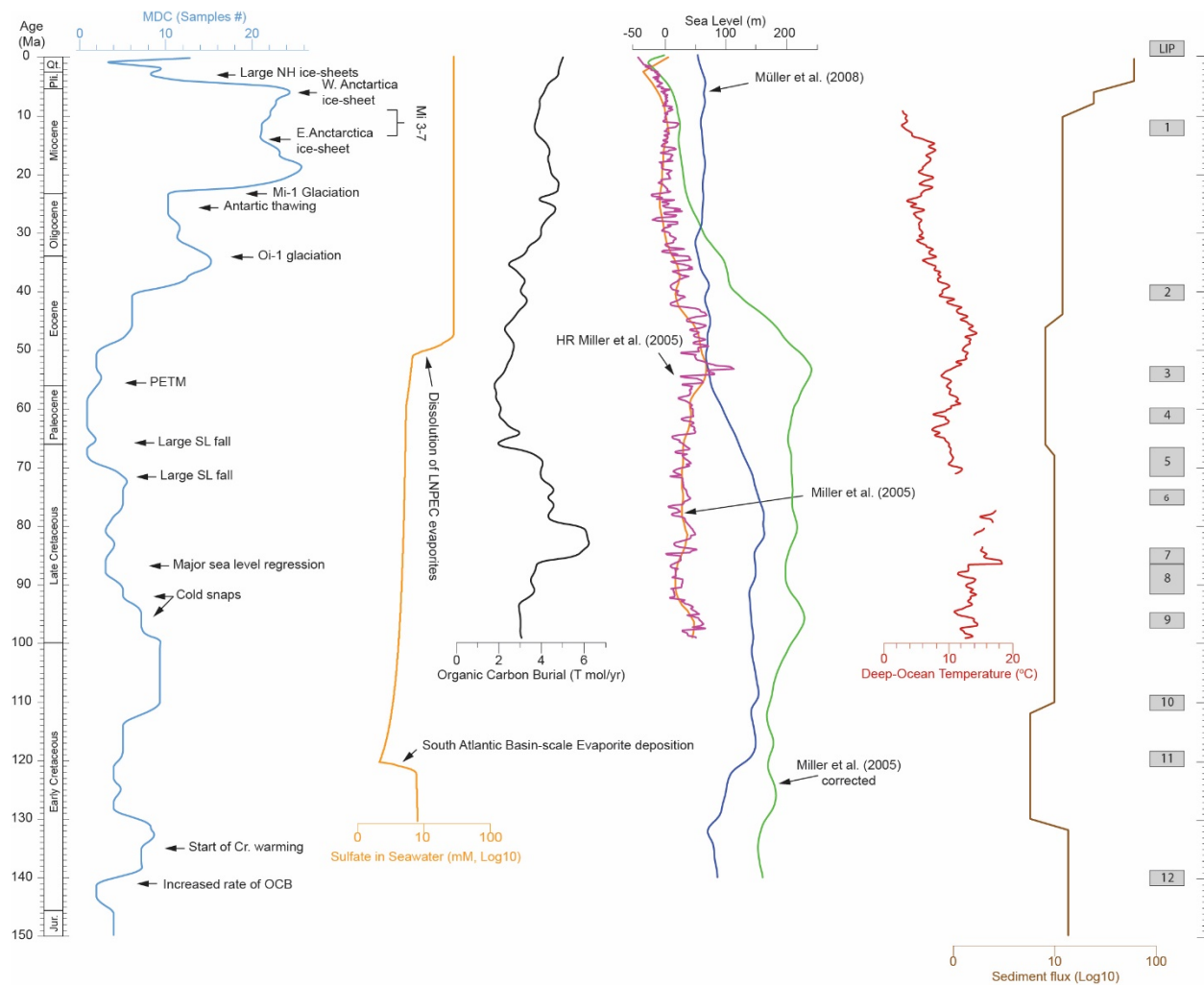
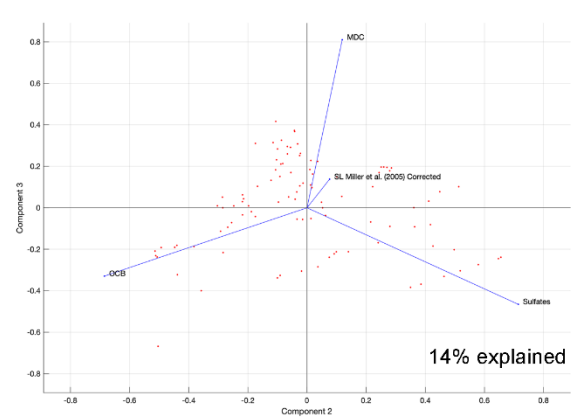
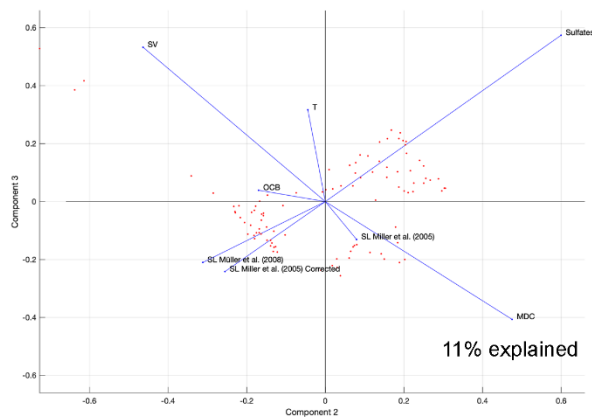
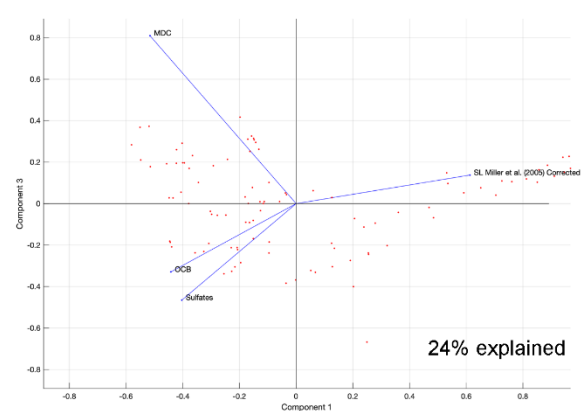
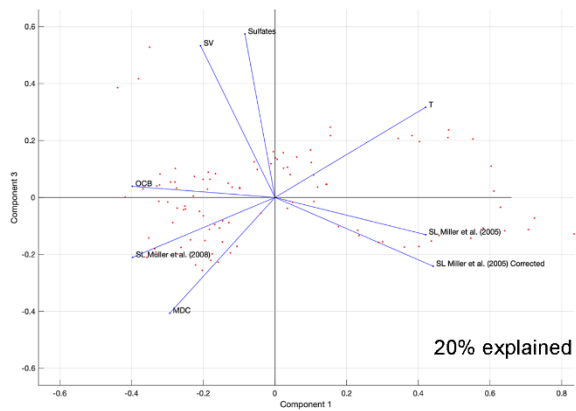
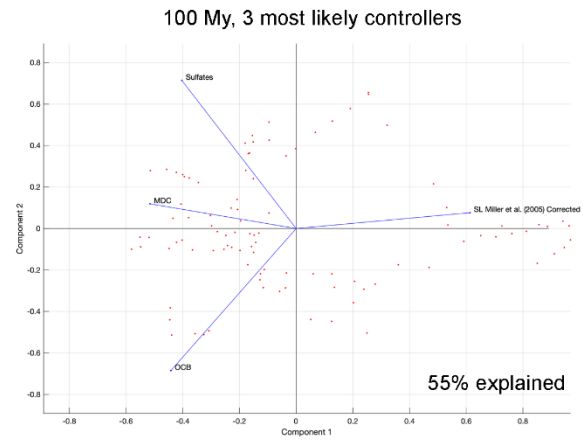
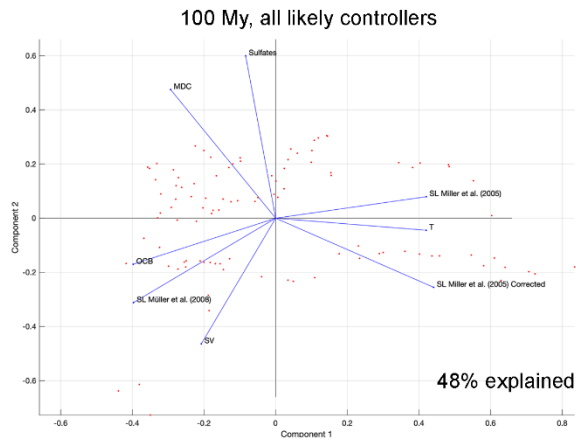


Fig. S1

Time series of methane-derived carbonates during the last 150 My. Records of Organic Carbon Burial (OCB), global sea level, deep-sea water temperature, seawater sulfate, sediment flux, and large igneous provinces (LIP) are also shown for comparison. The main climatic events are shown. References for the various time series are in the main text.



a)

b)

Fig. S2: Principal Component analysis. a) Principal component analysis performed on original data without regressions, on the ensemble of available measurements and models. The panels show the contribution of each observation in the two main planes defined by the first three components. These account for more than 79% of the total variance (PC1=48%; PC2=20%; PC3=11%). SV: sediment volume, SL: sea level, T: temperature, OCB: organic carbon burial, MDC: methane derived carbonate. b) Second Principal Component Analysis to assess the dependence of our inferences on the number of observations included. In this case we only consider the 4 most relevant observations.

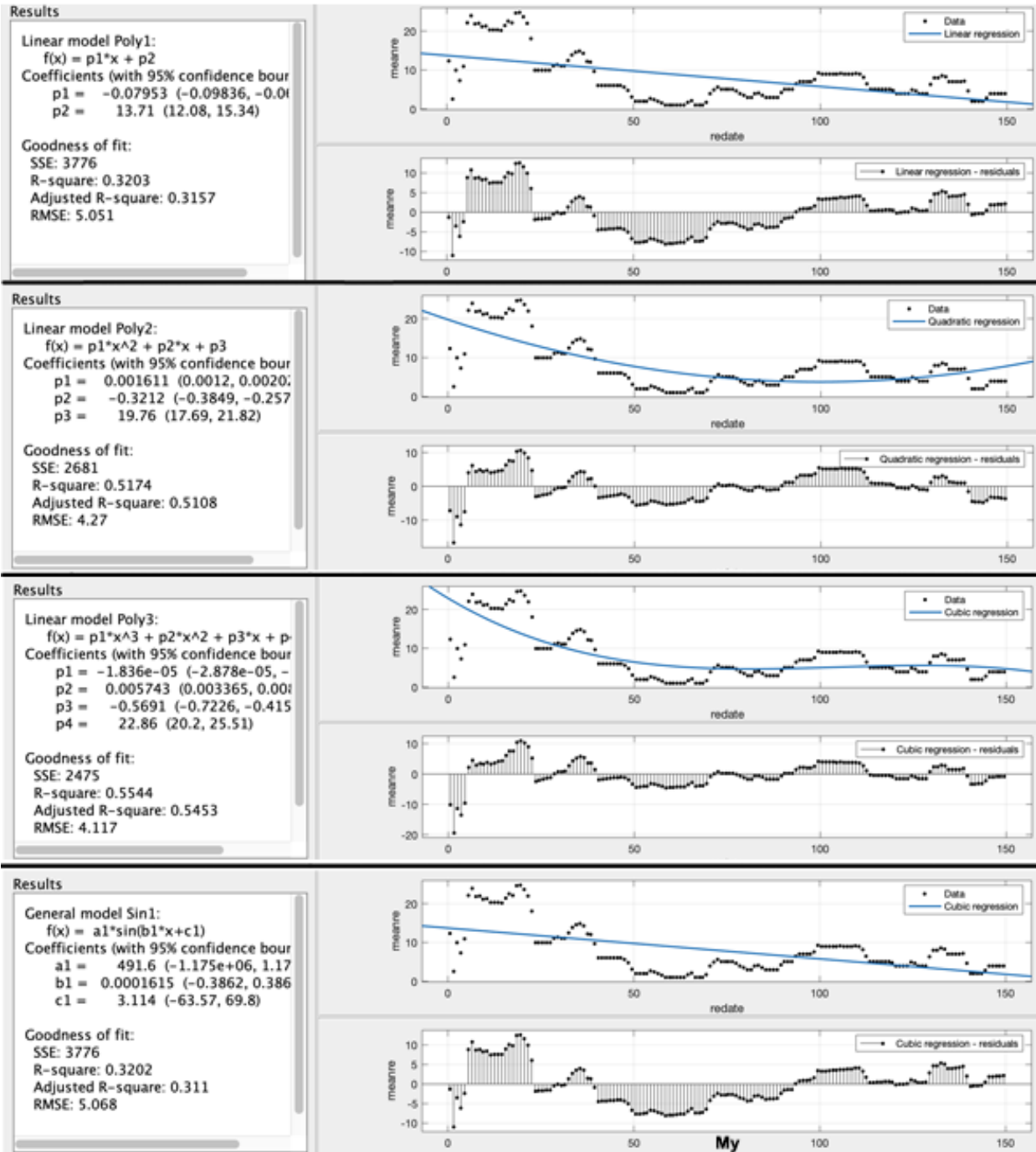


Fig. S3: Linear, quadratic, cubic and sinusoidal regressions with residuals.

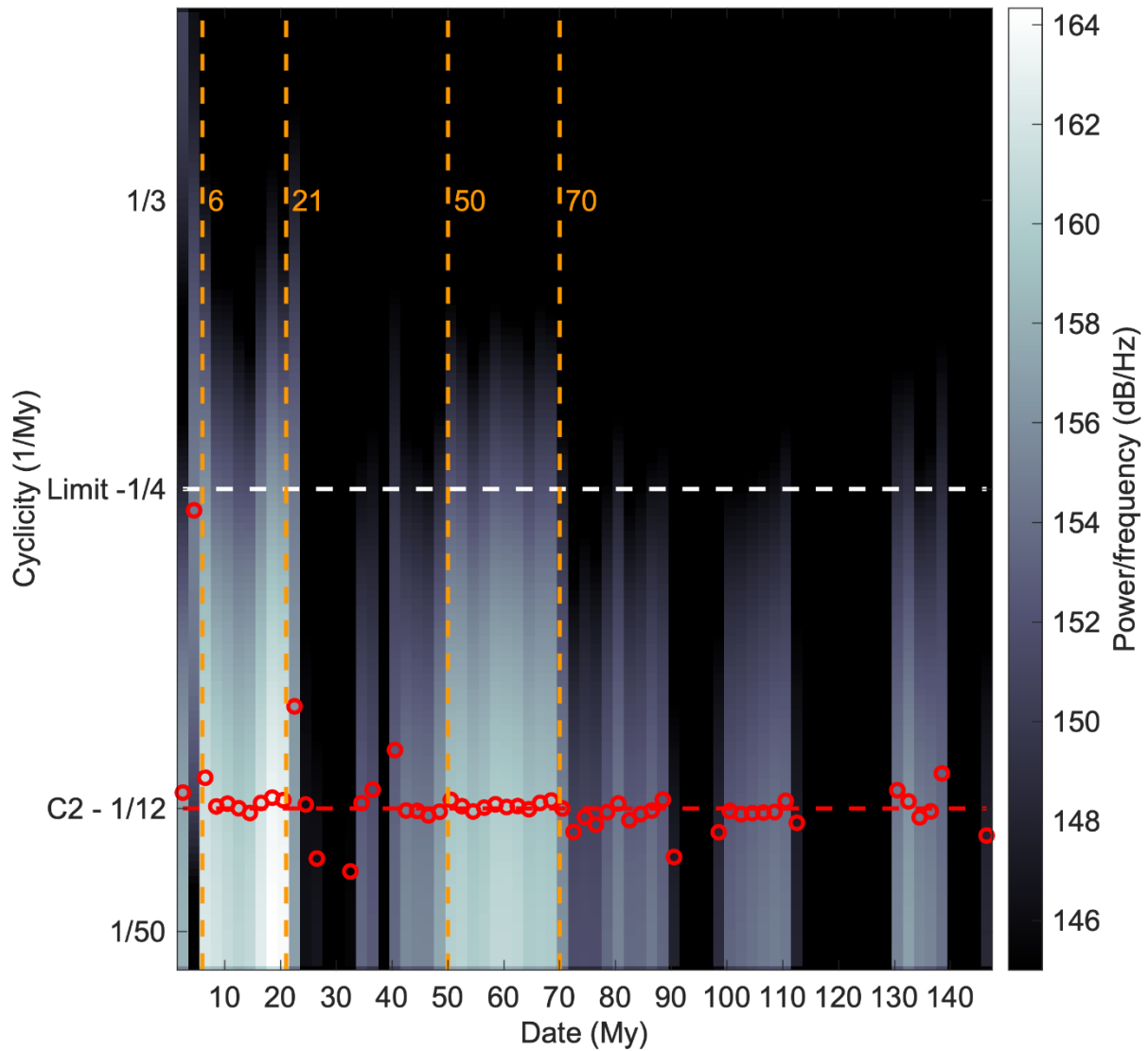
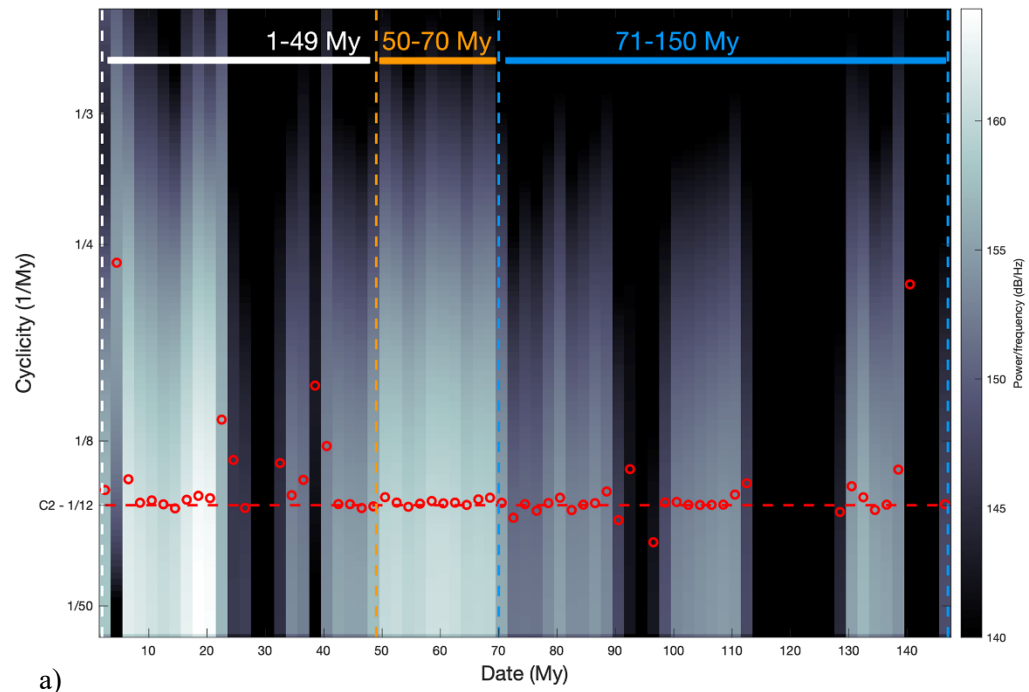
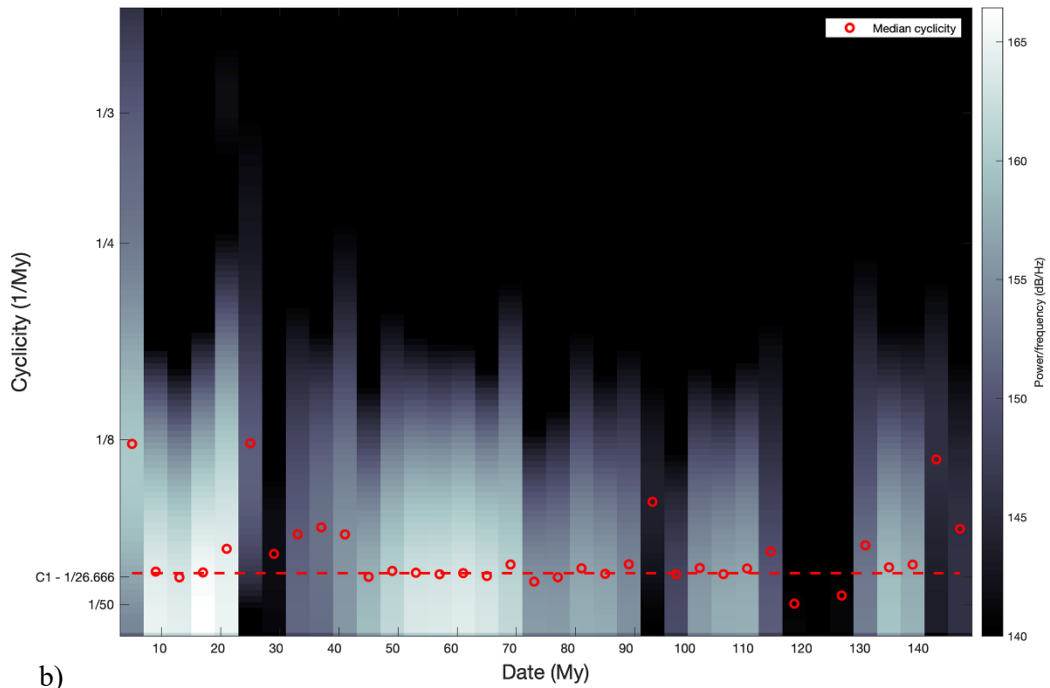


Fig. S4: Matched filtering of the MDC residuals using a Hamming windows of 5 My and 60% overlap. Threshold at 145 dB/My. The orange dashed lines indicate the period of the strongest MDC variations, characterise by the onset of a 1/12 cyclicity (C2 – red dashed line following the median across existing signals).



a)



b)

Fig. S5: Spectrograms of the MDC residuals computed using Hamming windows of 10 My and 5 My (60% overlap, threshold at 145 dB/My^{-1}). Red circles are median cyclicities. a) 5 My window again corresponding to median cyclicity C2; b) 10 My window corresponding to median cyclicity C1.

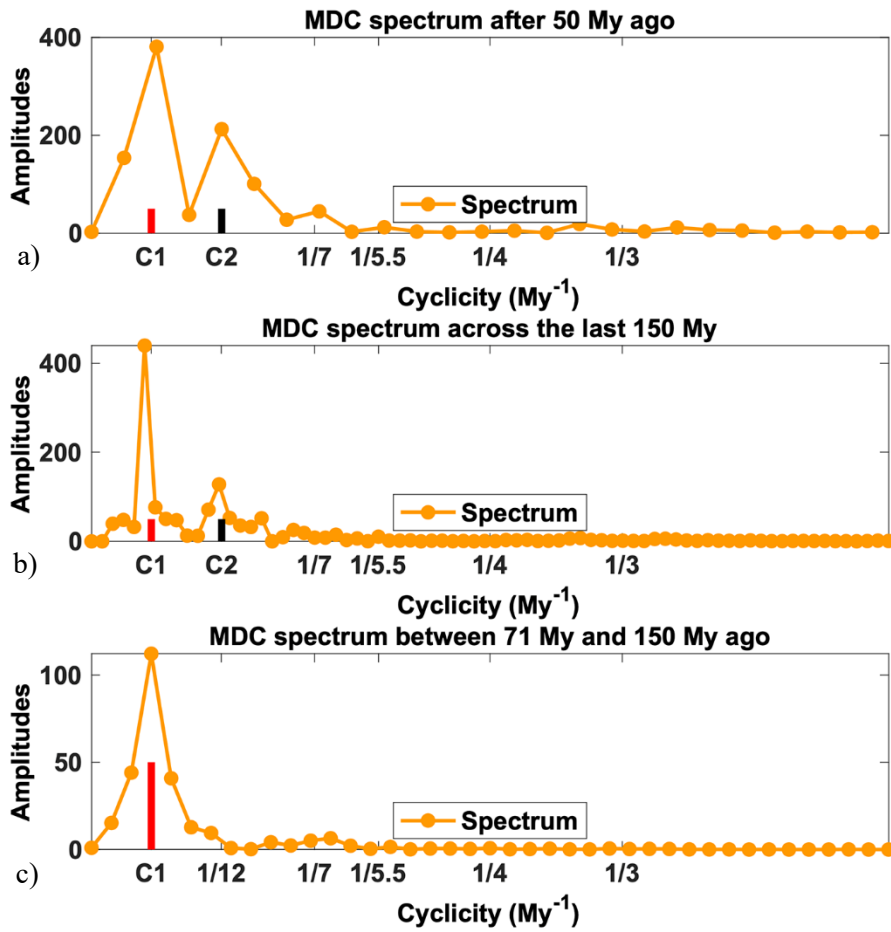


Fig. S6: Power spectrum of the MDC high-pass filtered residuals across the periods 1-49 Ma (a), 1-150 Ma (b), and 71-150 Ma (c). Cyclicity C2 does not appear in the bottom panel.

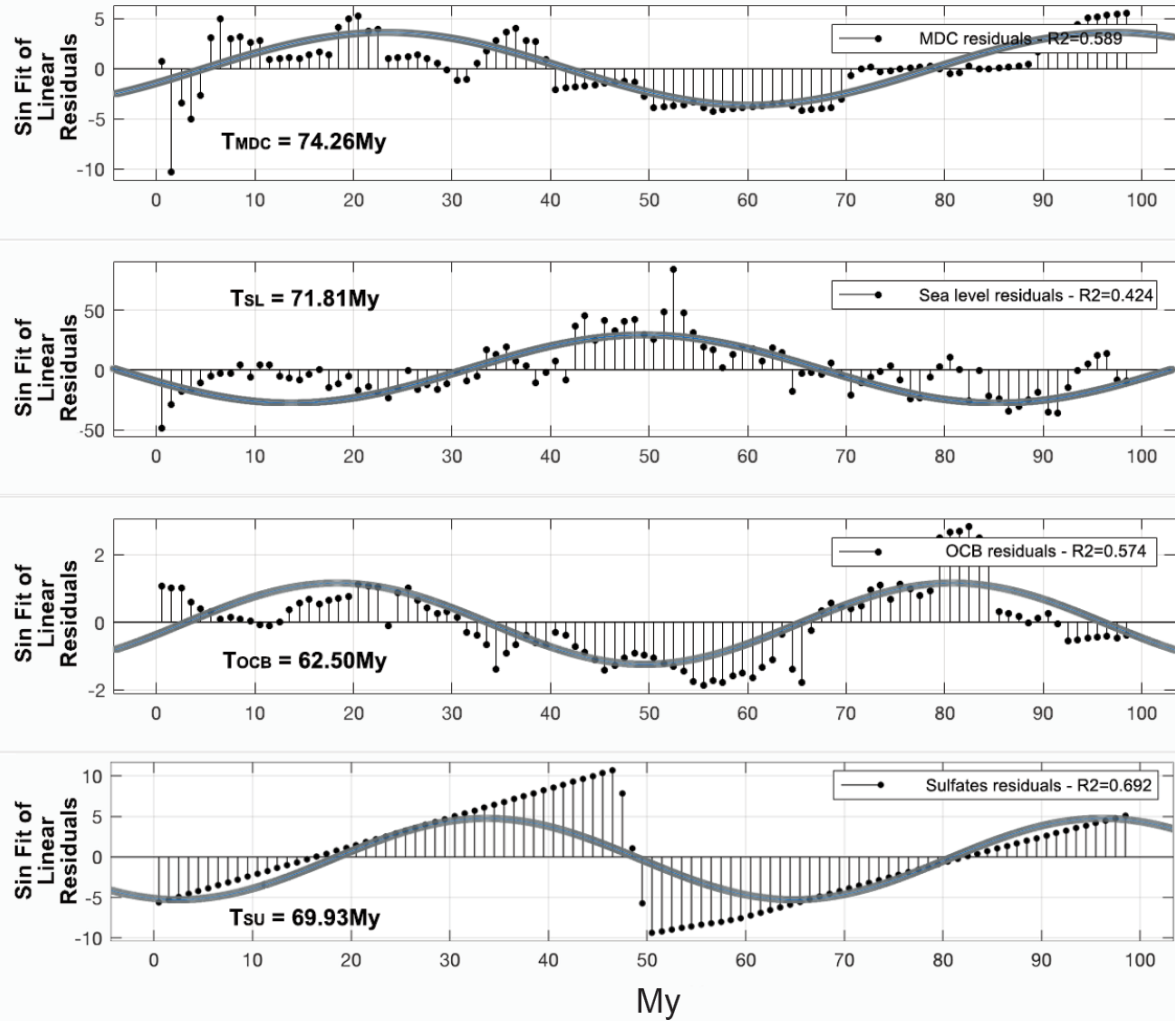


Fig. S7: Similar oscillatory signals characterising the linearly detrended MDC, sea-level, OCB and sulfates records.

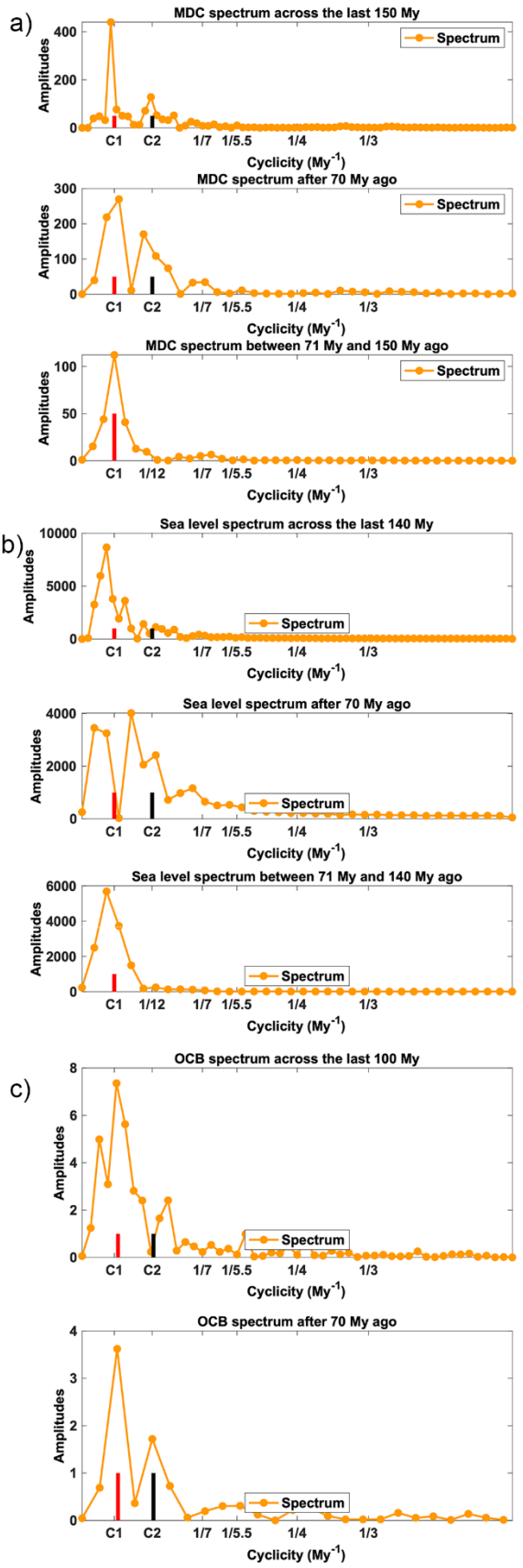


Fig. S8: Power spectrum of the MDC (a), sea-level (b) and OCB (c) high-pass filtered residuals across the periods: (a) 1-150 Ma (top), 1-70 Ma (centre), and 71-150 Ma (bottom); (b) 1-140 Ma (top), 1-70 Ma (centre), and 71-140 Ma (bottom); (c) 1-100 Ma (top), 1-70 Ma (bottom).

Supplementary Table 1. Seep-bearing Formations or equivalent

Age interval (Ma)	Formation name and/or location	Country/Location	Reference
0-0.120	Nile DSF	Mediterranean Sea	1
0-0.015	Tekirdag, Central & Cinarcik Basins, Marmara Sea	Turkey	2
0-0.120	Viking graben	North Sea	3
0.002-0.017	Offshore Norwegian margin	Norway	4
0.001-0.015	AT 340 site, Gulf of Mexico	USA	5
0.001-0.053	Alaminos Canyon, Gulf of Mexico	USA	5,6
0.001-0.005	Hydrate Ridge	USA	7
0.001-0.045	Green Canyon, Gulf of Mexico	USA	8
0.002-0.012	Hikurangi Margin	New Zealand	9
0.003-0.008	Offshore Svalbard	Svalbard	10
0.001-0.14	South China Sea	China	11
0.003-0.045	Hydrate Hole, offshore Congo Fan	Congo	12
0.001-0.005	Nile DSF	Mediterranean Sea	13
0.007-0.012	Amon mud volcano, NDSF	Mediterranean Sea	14
0.007-0.260	Hydrate Ridge	USA	7
0-0.010	Barkley Canyon	Canada	15
0.010-0.035	Offshore Joetsu	Japan	16
0.010-0.040	Offshore Noto Peninsula	Japan	17
0.015-0.019	Makran	Pakistan	18
0.021-0.023	Diapir field, offshore Congo Fan	Congo	12
0.030-0.060	Shenu area, South China Sea	China	19
0.032-0.033	Otago Peninsula	New Zealand	20
0.060-0.080	Offshore Gabon and Congo	W Africa	21
0-0.070	Mentawai fault zone	Java	22
0.001-0.047	Snail Hill, Sunda Arc	Java	22
0.250-0.360	Gulf of Cadiz	Portugal/Spain	23
0.137-0.143	Campos Basin	Brazil	24
0.250-0.270	Offshore Montenegro	Adriatic Sea	25
0.152-0.330	NE Dongsha	South China Sea	26
0.063-0.077	Shenhu	South China Sea	26
0.126-0.78	Kakinokidai Fm., Chiba Prefecture	Japan	27
0.118-0.138	Eel River Basin	USA	28
0-1	Monterey Bay	USA	29,30
0-1	Santa Barbara Basin	USA	29
0-1	Tommeliten	North Sea	29
0-1	Sea of Okhotsk	Russia	29
0-1	Kodiak Trench	USA	30
0-1	Nyegga area, Voring Basin	Norway	31
0-1	Codling Fault Zone	Irish Sea	32
0-1	Nile DSF	Mediterranean Sea	33

0-1	Offshore, St. Lawrence Estuary	Canada	34
0-1	Amsterdam mud volcano	Mediterranean Sea	35
0-1	Athina mud volcano	Mediterranean Sea	35
0-1	Offshore Costa Rica	Costa Rica	36
0-1	Bonaccia Gas Field	Adriatic Sea	37
0-1	Offshore eastern Aleutians	USA	38
0-1	Guaymas Basin	Mexico	39
0-1	Florida escarpment	USA	40
0-1	Makran accretionary prism	Pakistan	41
0-1	Outer Marianna Trough		42,43
0-1	Offshore Venice	Adriatic Sea	44
0-1	Krishna–Godavari Basin	India	45
0-1.8	Dnjepr canyon	Black Sea	46
1.4-1.65	Enza River, Northern Apennines	Italy	47
1.8-2.5	Koshiba Fm., Kanagawa Prefecture	Japan	27
1.8-3.6	Bata Fm	Philippines	48
1.8-23	Nadachi Fm.	Japan	49
2.5-3.6	Hijikata Fm., Shizuoka Prefecture	Japan	27
2.5-23	Quinault Fm	USA	50
2.5-3.6	Tamari & Horinouchi Fms., Shizuoka Prefecture	Japan	27
2.5-5.3	Quinault Fm; Quinault Coast	USA	51
2.5-5.3	Maarai Fm., Chiba Prefecture	Japan	27
2.5-5.3	Shiramazu Fm., Chiba Prefecture	Japan	27
2.5-5.3	Kawazume Fm., Niigata Prefecture	Japan	27
2.5-5.3	Quinault Fm., Washington	USA	15
3.6	Stirone River	Italy	52
3.6-28	Navidad Fm.	Chile	53
5-11	Santa Cruz Mudstone	USA	54
5-11	Hunghuatzu Fm., Nantzuhsien River	Taiwan	55
5-16	Montardone melange, N. Apennines	Italy	56
5-23	Columbia River, Washington	USA	57
5.3-11.6	Nodani Fm.	Japan	58
5.3-23	Taira Fm	Japan	59
5.3-23	Ogaya Fm.	Japan	60
5.3-23	Bessho Fm	Japan	60
5.3-23	Bexhaven Limenstone	New Zealand	61
5.3-23	Hiranita Fm.	Japan	62
5.3-33.9	Sagavanirktok Fm., North Slope Alaska	USA	63
5.3-11.6	Nodani Fm., niigata Prefecture	Japan	27
5.3-11.6	Morai Fm., Central Hokkaido	Japan	27
5.3-23.3	Moonlight limestone; NE Coastal basin Northern Island	New Zealand	64
5.3-23.3	Marmorito, Monferrato	Italy	65
5.3-7.2	Verrua Savoia, Tertiary Piedimont Basin	Italy	66

5.3-7.2	Ripa dello Zolfo, Tertiary Piedimont Basin	Italy	66
5.3-7.2	Calcare Solfitero, Sicily	Italy	67
5.4	Akaishi Fm., Fukaura Town	Japan	68
5.5-9.3	Perto Escondido	Venezuela	69
6-8	Lucina limestone	Italy	70
6-11.1	Godineau River	Trinidad	69
6.4-10	Jordan Hill	Trinidad	69
6.65-21.25	Caujarao	Venezuela	69
8	Lucina limestone, Pietralunga	Italy	65
8.2-9	Urenui Fm., Taranaki Basin	New Zealand	71
10-20	Hayama Fm	Japan	72
10-20	Lengua Fm	Trinidad	73
10-20	Pozon Fm	Venezuela	73
11-16	Lucina limestone	Italy	70
11.6-23	Higashibessh Fm.	Japan	74
11.6-23	Fosso Riconi	Italy	75
11.6-16	Nupinai Fm.	Japan	76
13.3-13.75	Vicchio Marls	Italy	77
15	Hayama Fm.	Japan	78
15.9-23	Setogawa Group, Shizuoka Prefecture	Japan	27
15.9-23	Aokiyama Fm., Chiba Prefecture	Japan	27
16-20.4	Kokozura Fm.	Japan	79
16-23	Muroto Fm	Japan	80
16-23	Shikiya Fm.	Japan	81
16.85-17.3	Plamar-Molinera	Colombia	69
16.85-17.3	Plamar-Molinera	Colombia	69
18.75-23.3	Bath Cliff	Barbados	69
18.5-20	Cantera portugaleta	Cuba	69
18.6-21.25	Corro Colorado	Venezuela	69
20-30	Astoria Fm	USA	82
20-30	Talara Fm	Peru	83
20-30	Heath shales	Peru	83
20.5-23.1	Buena Vista de Maicillal	Venezuela	69
23-28	Pysht/Sooke Fm., Washington	USA	15
23-28	Keasey Fm	USA	84
23-33	Krosno Fm	Poland	85
23-33	Lincoln Creek Fm	USA	86
23-33	Satsop River, Lincoln Creek	USA	57
23-33	Clatskanie, Oregon	USA	57
28-33	Makah Fm., Washington	USA	57
28-33.9	Heath shales, Belén	Peru	87
28.4-33.9	Nuibetsu Fm., Kami-Atsunai Station	Japan	68
28.4-37.2	Tanamigawa Fm., Honshu	Japan	80

30-36	Canyon River, Washington	USA	88
30-40	Diapiric melange	Barbados	89
30-40	SOFZ	Barbados	89
33.9-55.8	Poronai Fm., Central Hokkaido	Japan	27
33-37	Wagonwheel Fm	USA	83
33.9-48	West Fork of Grays River, Washington	USA	88
33-40	Humptulips Fm	USA	86
33-40	Lincoln Creek Fm	USA	86
33.9-40	Bear River, Washington	USA	88
33-40	Siltstone of Units B	USA	86
33-40	Whiskey Creek	USA	90
33-40	Humptulips Fm	USA	91
33-55	Keasey Fm., Oregon	USA	57
35-49.25	Elmira asphalt mine	Cuba	69
36.3-37.6	Joes River	Barbados	69
40-50	Central Istria Flysh	Croatia	92
40-50	Oomagari Fm	Japan	93
50	Pobiti Kamani	Bulgaria	94
55-58	Fossildalen	Svalbard	95
55-65	Moreno Fm, Panoche Hills	USA	19,96
65-66	James Ross Basin, Snow Hill Island	Antarctica	97
65-66	James Ross Basin, Seymour Island	Antarctica	88
65.5-99.6	Sada Limestone, Shikoku	Japan	98
70.3-73	Tepee Buttes	USA	99,100
70.6-83.5	Guenoc Ranch & Romero Creek	USA	101
70-80	Omagari Fm	Japan	102
72-126	Raukumara Peninsula	New Zealand	103
72.1-78	Western Interior Seaway, Montrose	USA	88
78	Whangai Fm. North Island	New Zealand	103
83-85	Maeshima, Kyushu	Japan	88
83-100	Shangba Section	Tibet	104
90-100	Tenkaritoge Fm	Japan	105
90-100	Kadui, Jiding, and Walie Sections	Tibet	104
93.6-99.6	Ezo Group, NW Hokkaido	Japan	27
94-100	Gamba Section	Tibet	104
99.6-112	Ezo Group, Central Hokkaido	Japan	27
99.6-125	Great Valley Group, Cold fork of Cottonwood	USA	106
99-112	Ispaster	Spain	107
100-110	Hikagenosawa Fm	Japan	108
100-113	Black Flysch Group, Basque-Cantabrian Basin	Spain	109
100-113	Ellef Ringnes Island	Canada	110
100-113	Xiege Section	Tibet	104
100-125	Christopher Fm	Canada	111

106	Yezo Group	Japan	78
110-120	Logoda Fm	USA	84
113-120	Marnes Bleues Fm., Vocontian Basin	France	112
120-130	La Perlita limestone	Mexico	84
125-130	Kuhnpasset Beds	Greenland	113
125-130	Hradiste Fm., Baska	Czech Republic	114
125-134	Sinaia Fm, Carpatian Mountains	Romania	115
130-133.9	Great Valley Group, Wilburg Spring & Rice Valley	USA	106
130-134	Planerskoje section	Ukraine	116
130-140	Chebo Fm	China	84
130-140	Maiolica Fm	Italy	84
130-140	Sangxiu Fm	China	84
130-140	Upper Grodziszczce Fm	Czech Republic	84
130-134	Curnier, Rottier	France	117
133.9-140	Great Valley Group, Rocky & Bear Creeks	USA	118
133-140	Crack Canyon Fm, Rocky Creek	USA	118
134-139	Sandy limestone, Novaya Zemlya	Russia	119
139-152	Sassenfjorden	Svalbard	120
140-150	Stony Creek Fm	USA	84
145	Black Limestone, Novaya Zemlya	Russia	119
145.5-150.8	Great Valley Group, Paskenta	USA	106
145-150	Gateway Pass beds	Antarctica	121

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