

Data Repository Item: Appendix DR1 Model treatment of seawater cation composition

Analysis of fluid inclusions contained in marine halite crystals reveals a substantial Phanerozoic variability in magnesium (Mg^{2+}) and calcium (Ca^{2+}) ion concentrations (Lowenstein et al., 2001; Horita et al., 2002; Lowenstein et al., 2003). This will affect global carbonate cycling and the sedimentary buffering of atmospheric pCO_2 , firstly because $[Ca^{2+}]$ directly determines the solubility ratio of calcite: $\Omega = [Ca^{2+}] \times [CO_3^{2-}] / K_{sp}$. Changes in the concentration of magnesium ions also influences the solubility of calcite as well as modifying the stoichiometric equilibrium constants. The aqueous carbonate chemistry model of Ridgwell et al. (2007) has therefore been extended to capture the 1st-order importance of these effects.

The solubility coefficient for calcite was modified following Tyrrell and Zeebe (2004):

$$K_{sp}^* = K_{sp,modern}^* - \alpha \cdot \left(\frac{[Mg^{2+}]_{modern}}{[Ca^{2+}]_{modern}} - \frac{[Mg^{2+}]}{[Ca^{2+}]} \right) \quad \text{Eq. DR1-1}$$

where $K_{sp,modern}^*$ is the solubility coefficient of calcite for a modern seawater composition (Mucci, 1983), $[Mg^{2+}]_{modern}$ and $[Ca^{2+}]_{modern}$ are the modern mean seawater concentrations of Mg^{2+} and Ca^{2+} of 52.82 and 10.25 mmol/kg, respectively and $\alpha = 3.655 \times 10^{-8}$. Early Eocene (55 Ma) $[Mg^{2+}]$ and $[Ca^{2+}]$ values of 29.9 and 18.2 mmol/kg, respectively, were taken from the empirical Cenozoic curve of Tyrrell and Zeebe (2004).

The tendency of Mg²⁺ to form ion pairs in seawater (Zeebe and Wolf-Gladrow, 2001) requires that the 1st and 2nd equilibrium constants of carbonic acid, K_1^* and K_2^* are adjusted:

$$K^* = \left(1 + s_{K^*} \cdot \frac{[\text{Mg}^{2+}] - [\text{Mg}^{2+}]_{\text{modern}}}{[\text{Mg}^{2+}]_{\text{modern}}} \right) \cdot K_{\text{modern}}^* \quad \text{Eq. DR1-2}$$

where K_{modern}^* is the modern seawater composition value (Mehrbach et al. (1973), as refined by Dickson and Millero (1987)) and s_{K^*} is a sensitivity parameter. The values of s_{K^*} for K_1^* and K_2^* are 0.155 and 0.442, respectively (Ben-Yaakov and Goldhaber, 1973).

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Data Repository Item: Appendix DR2
Paleode

p_{th} and CaCO₃ content data used in this study

Sites in data-rich regions are indicated by site numbers in **boldface**.

Site	<u>Paleodepth</u>		<u>Sediment CaCO₃ wt%</u>		
	Depth (m)	Source	Baseline	PETM	Source
<u>Pacific Ocean</u>					
163	4490	Van Andel (1975)	absent	—	Van Andel (1975)
164	5220	Van Andel (1975)	hiatus*	—	Van Andel (1975)
165	4570	Van Andel (1975)	absent	—	Van Andel (1975)
166	4880	Van Andel (1975)	hiatus*	—	Van Andel (1975)
168, 169	5000	Van Andel (1975)	hiatus*	—	Van Andel (1975)
170	5250	Van Andel (1975)	hiatus*	—	Van Andel (1975)
303	5540	Van Andel (1975)	hiatus*	—	Van Andel (1975)
304	5600	Van Andel (1975)	hiatus*	—	Van Andel (1975)
307	5520	Van Andel (1975)	hiatus*	—	Van Andel (1975)
313	3200	Van Andel (1975)	present	—	Van Andel (1975)
315	4120	Van Andel (1975)	hiatus*	—	Van Andel (1975)
316	4020	Van Andel (1975)	hiatus*	—	Van Andel (1975)
577	1900	Miller et al. (1987)	96	hiatus	Bralower et al. (2002), Quillévéré et al. (2002)
865	1300 – 1500	Thomas & Shackleton (1995)	96	93	Thomas et al. (1999), SSP (1995) [§]
1209	2000 – 2500	SSP (2002a) [§]	96	84	Colosimo et al. (2006)
1210	2000 – 3000	SSP (2002b) [§]	92	86	Colosimo et al. (2006)
1211	2000 – 3000	SSP (2002c) [§]	95	78	Colosimo et al. (2006)
1212	2000 – 3000	SSP (2002d) [§]	98	83	Colosimo et al. (2006)
1215	3200	Rea and Lyle (2005)	74	—	SSP (2002e) [§]
1217	3200	Rea and Lyle (2005)	present	—	SSP (2002f) [§]
1220	2900	Rea and Lyle (2005)	90	0	SSP (2002g) [§]
1221	3200	Rea and Lyle (2005)	74	3	Murphy et al. (2006)
LL4-GPC3	—	—	absent	—	Kyte et al. (1993)
<u>Caribbean Sea</u>					
999	1750	Thomas et al. (1999)	61	0	Bralower et al. (1997)
1001	1500	Thomas et al. (1999)	45	0	Bralower et al. (1997)
<u>Atlantic Ocean</u>					
527	3400	Moore et al. (1984)	83	0	Thomas et al. (1999)
549	2000 – 2300	Masson et al. (1985)	51	1	Thomas & Bralower (2005)
689	1400 – 1650	Kennett & Stott (1991)	91	—	SSP (1988) [§]
690	2100	Zachos et al. (1993)	84	60	Thomas et al. (1999)
1050	—	—	63	57	Rudnicki et al. (2001)
1051	1000 - 2000	SSP(1998) [§]	56	52	Rudnicki et al. (2001)
1262	3600	Zachos et al. (2005)	88	1	Zachos et al. (2005)
1263	1500	Zachos et al. (2005)	88	1	Zachos et al. (2005)

1266	2600	Zachos et al. (2005)	85	4	Zachos et al. (2005)
1267	3200	Inferred [†]	80	1	Zachos et al. (2005)

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Paleodepth and CaCO₃ content data: continued

Site	Paleodepth		Baseline	Sediment CaCO ₃ wt%	
	Depth (m)	Source		PETM	Source
Indian Ocean					
211	4900	Van Andel (1975)	absent	—	Van Andel (1975)
212	6050	Van Andel (1975)	hiatus*	—	Van Andel (1975)
213	3520	Van Andel (1975)	present	—	Van Andel (1975)
215	3450	Van Andel (1975)	present	—	Van Andel (1975)
235	4300	Van Andel (1975)	absent	—	Van Andel (1975)
236	2740	Van Andel (1975)	present	—	Van Andel (1975)
239	4220	Van Andel (1975)	absent	—	Van Andel (1975)
240	3200	Van Andel (1975)	present	—	Van Andel (1975)
241	3990	Van Andel (1975)	absent	—	Van Andel (1975)
245	3560	Van Andel (1975)	present	—	Van Andel (1975)
248	4170	Van Andel (1975)	hiatus*	—	Van Andel (1975)
250	5080	Van Andel (1975)	hiatus*	—	Van Andel (1975)
256	5000	Van Andel (1975)	hiatus*	—	Van Andel (1975)
259	4000	Hancock et al. (2006)	61	37	Hancock et al. (2006)
260	5080	Van Andel (1975)	hiatus*	—	Van Andel (1975)
261	5400	Van Andel (1975)	hiatus*	—	Van Andel (1975)
738	1350	Barrera & Huber (1991)	90	70	SSP (1989) [§]
766	3476	SSP (1990) [§]	93	—	SSP (1990) [§]
1135	1000 – 2000	Inferred [‡]	95	—	SSP (2000) [§]
1138	600 – 1200	Quilty (2002)**	93	—	SSP (2000) [§]

* Due to non-deposition

§ SSP = Shipboard Scientific Party

† At present, Site 1262 is 400 m deeper than 1267. To estimate a paleodepth for 1267, 400 m was subtracted from the paleodepth of Site 1262.

‡ Inferred from paleodepths of surrounding sites, including the Paleocene paleodepths of Sites 738 (Barrera and Huber, 1991), 747, and 748 (Mackensen and Berggren, 1992), and Late Maastrichtian paleodepths of Sites 747, 748, 750 (Quilty, 1992), and 1138 (Quilty, 2002), and given the bathymetric divisions of Berggren and Miller (1989).

** Late Campanian to Late Maastrichtian paleodepth.

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**Data Repository Item: Appendix DR3:
Evaluation of goodness-of-fit between model output and observations**

We address uncertainty regarding the Late Paleocene global weathering rate and the ratio of CaCO_3 to particulate organic carbon in the export of biogenic detritus from the surface ocean (the CaCO_3 :POC rain ratio) by performing a 2-parameter sensitivity analysis comprising an ensemble of simulations with weathering fluxes of 15, 20, 25, 30, 35, and 40 Tmol a^{-1} of HCO_3^- and spatiotemporally uniform rain ratios of 0.150, 0.175, 0.200, 0.225, and 0.250. Each ensemble member was spun up for 150 ka to reach geochemical equilibrium between weathering and sedimentation (Fig. DR3-1).

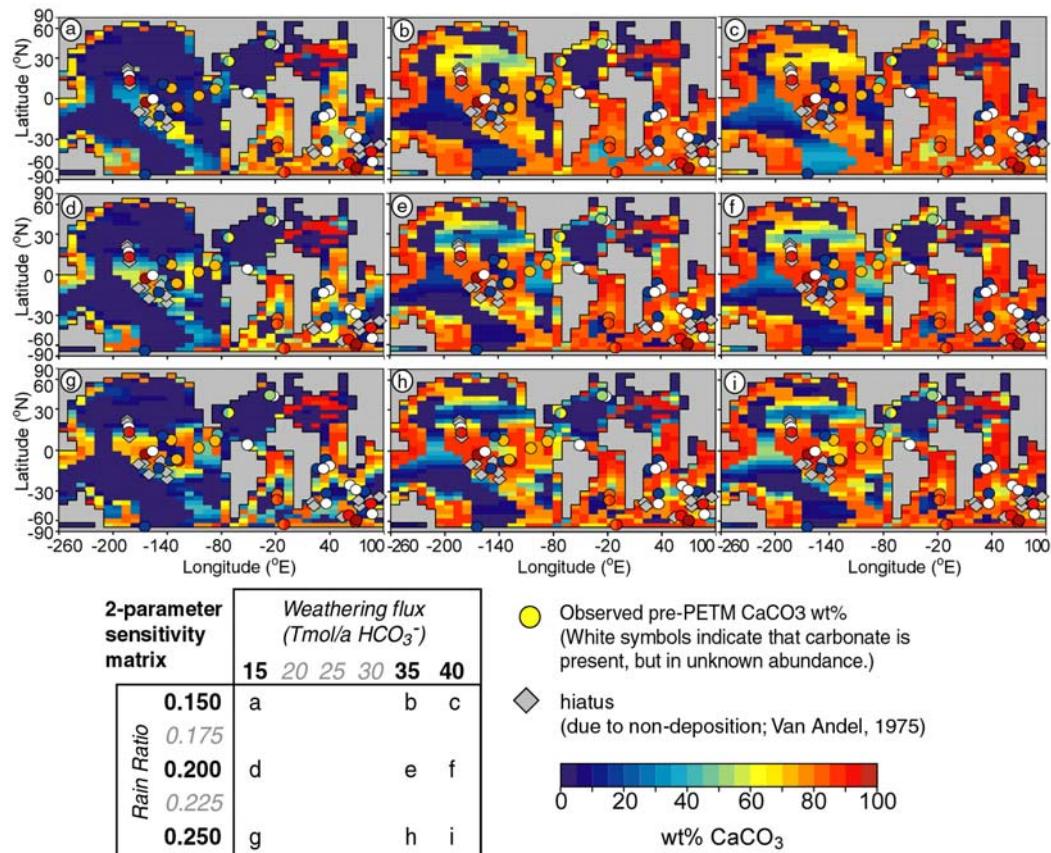


Figure DR3-1: Coretop CaCO_3 wt% output (maps) for a subset of model runs (panel labels correspond to the table in the legend) with observations superimposed (circle and diamond markers).

The best fit between model output and observed CaCO₃ wt% was identified by finding the ensemble member with the lowest root mean squared error (RMSE) between model output and observations,

$$RMSE = \sqrt{\frac{1}{n} \sum (result - observation)^2} \quad \text{Eq. DR3-1}$$

where n is the number of comparisons. First, CaCO₃ wt% was plotted versus depth for each of the model outcomes and the observations for three relatively data-rich regions: the central equatorial Pacific, Walvis Ridge in the Atlantic Ocean, and the southern Indian Ocean (identified as a, b, and c in Fig. 1A in the text; see DR2 for data and citations). The profiles were fit using a linear weighted regression scheme described by Cleveland and Devlin (1988), and available on-line in a data visualization toolbox (version 1.1) from Datatool (www.datatool.com) as the script loess.m for Matlab. This script requires that the user specify a set of points at which to compute the value of the fitted function, here set at 200 m increments; a smoothing parameter, α ; and the order of the polynomial to be used by the Matlab function polyfit, λ .

Next, the calculated fits to both model output and observations were evaluated at model layer depths encompassed by the paleodepths of observations. The RMSE was calculated for each region (Fig. DR3-2, A,B,C) and the total RMSE was computed (Fig. DR3-2D).

The best overall model-data fit (i.e., the scenario with the lowest total RMSE) is obtained with a CaCO₃:POC rain ratio of 0.200, higher than the mean value of ~0.14 characterizing the modern ocean (Ridgwell et al., 2007), and a total global weathering rate of 35 Tmol/a of HCO₃⁻, which is within the range of the modern estimate,

23-39 Tmol/a of HCO₃⁻ (Munhoven, 2002) but less than that from early Eocene model simulations (Gibbs et al., 1999). The best overall model-data fit has a global average CaCO₃ wt% of 50% (Fig. DR3-1E).

The same analysis was performed to evaluate the model scenario with the best fit to observed PETM CaCO₃ wt% distributions (Fig. 4 in the text), except that the southern Indian Ocean region was not included because there are insufficient observations to generate a meaningful fit.

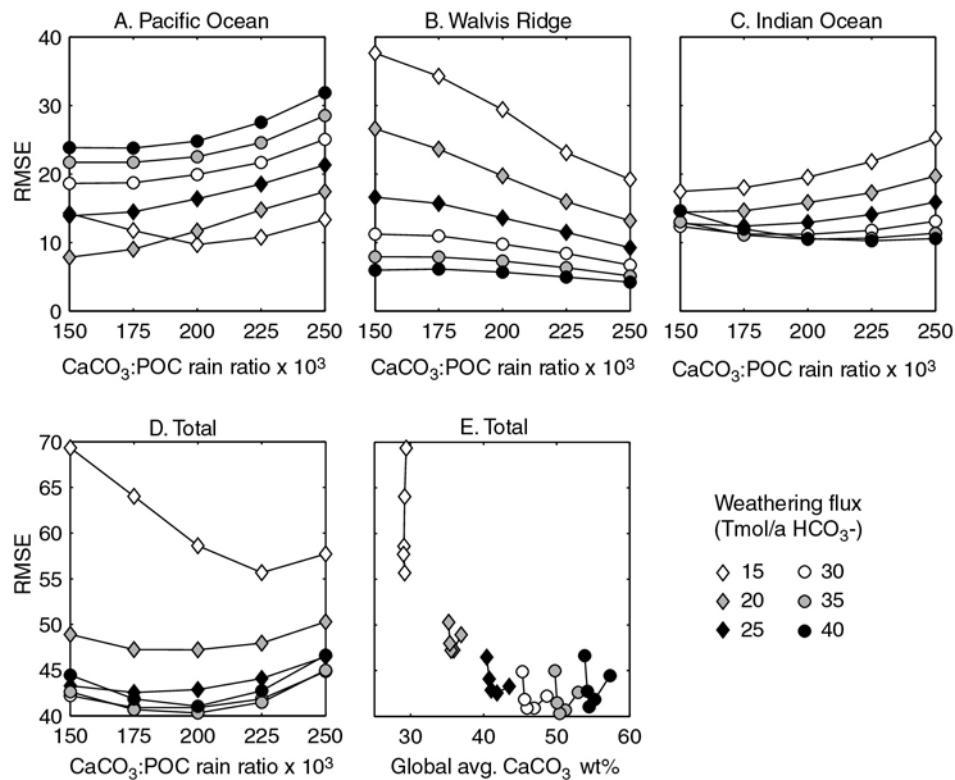


Figure DR3-2: Root mean squared errors (RMSE) for the weathering flux and rain ratio sensitivity ensemble. The lowest overall RMSE (D, the sum of RMSE for data-rich regions in A, B, and C) is achieved with a weathering flux of 35 Tmol/a of HCO₃⁻ and a rain ratio of 0.200. This scenario corresponds to a global average model CaCO₃ wt% of 50% (E).

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Data Repository Item: Appendix DR4
Comparison of model and observed bottom water $\delta^{13}\text{C}$ latitudinal gradient

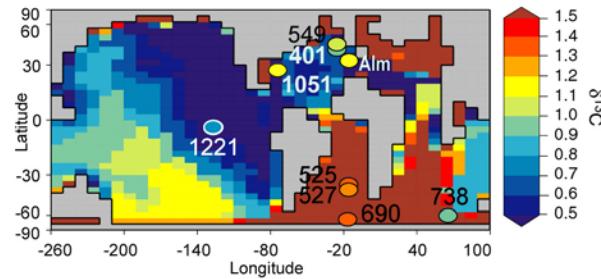


Figure DR4-1: Pre-PETM model bottom water $\delta^{13}\text{C}$ with measurements of benthic foraminiferal $\delta^{13}\text{C}$ as compiled by Nunes and Norris (2006) superimposed (circular markers).

Table DR4-1: Model and observed pre-PETM bottom water $\delta^{13}\text{C}$ values

Basin	Site	Bottom water $\delta^{13}\text{C}$		Paleolatitude (°N)
		Model	Data: benthic forams*	
Pacific Ocean	1221	0.4	1.1	-5.8
Atlantic Ocean	690	2.1	1.6	-66.2
	525	1.9	1.3	-36.8
	527	1.9	1.1	-31.6
	1051	0.4	1.3	27.9
	ALM	0.5	0.9	31.9**
	401	0.7	1.0	42.1
	549	0.7	1.1	43.5
Indian Ocean	738	2.1	0.9	-60.9

* Nunes and Norris (2006)

** Picked from model grid

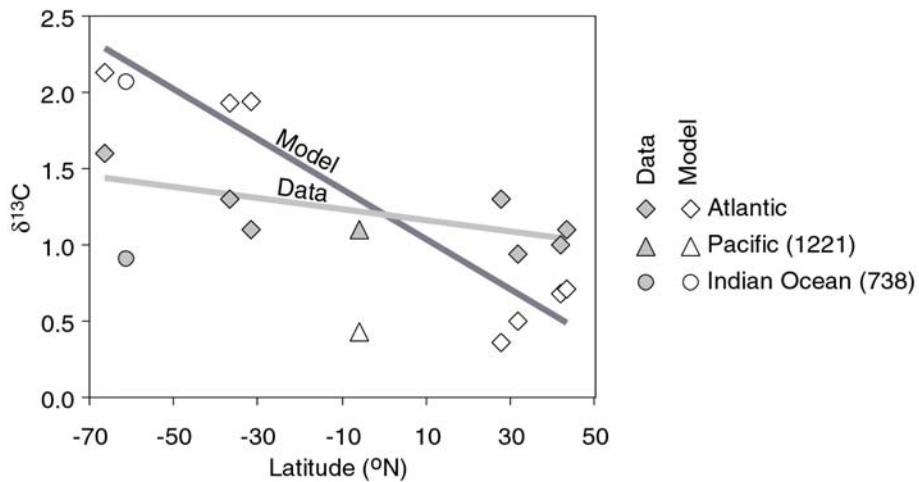


Figure DR4-2: Model and observed latitudinal gradient in bottom water $\delta^{13}\text{C}$. Observations from Nunes and Norris (2006).

References

Nunes, F., and Norris, R.N., 2006, Abrupt reversal in ocean overturning during the Palaeocene/ Eocene warm period: Nature, v. 439, p. 60-63.