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Silicate weathering and North Atlantic silica burial during the Paleocene-Eocene Thermal Maximum

Donald E. Penman

Parameter	**Value**	**Reference**
Surface box volume (m^3)	3.58E+16	
Deep box volume (m^3)	1.30E+18	
Height of surface box (m)	100	
Hydrothermal input flux (mol Si/yr)	6.00E+11	*a*
Aeolian (dust) input flux (mol Si/yr)	5.00E+11	*a*
Background riverine input flux (mol Si/yr)*	5.60E+12	*a*
Surface-deep mixing rate (m/yr)	3	*b*
fraction of opal production remineralized in surface(%)	50	*a*
fraction of opal production remineralized in deep (%)*	47.42	*a*
Opal burial fraction (%)*	2.58	*a*
Background surface opal production (mol Si/yr)*	2.60E+14	*a*

Table DR1: Parameters describing the 2-box silica cycle model used in this study. See main text Figure 1 for model architecture. Values that are manipulated in various versions of PETM experiments are denoted with an asterix (*) and described in the text below. References: *a*: the silica cycle budget of Sarmiento and Gruber (2006), based on Treguer et al. (1995), Nelson et al. (1995) and DeMaster (2002), *b*: Broecker and Peng (1982).

PETM forcing: To simulate the sequestration of 5,000 or 10,000 GtC by silicate weathering over 150kyr (see main text), the riverine Si input is increased from 5.6 to (respectively) 8.4 or 11.2 Tmol Si/year for 150 kyr and then returns to 5.6 Tmol/Si/year for the remainder of the run.

Surface opal production: The uptake of dissolved silica in the surface ocean by siliceous plankton is parameterized by two alternative functions of surface dissolved silica concentration ($[H_4SiO_4]$), providing a stabilizing feedback to changes in $[H_4SiO_4]$.

The first (denoted "linear uptake" in Fig S1) is a linear function going through the origin (no uptake when [H_4SiO_4] = 0) and a point defined by modern global uptake at average modern [H_4SiO_4] (260 Tmol/year when [H_4SiO_4] = 7.7mmol/m^3):

$$F_{OPAL} = [H_4SiO_4]_{SURF} / .0077 * 260 \text{ Tmol Si/year}$$

This parameterization provides a strong negative feedback to changes in [H_4SiO_4], but assumes that the availability of dissolved silica is the only limitation on populations of silicifiers, i.e. that their abundance can increase boundlessly if [H_4SiO_4] is high enough. In the real world, they are also limited by other factors, importantly N and P availability for diatoms (Yool and Tyrrell, 2003) and food supply for radiolarians. To simulate these limitations, a second parameterization of silica uptake (denoted "asymptotic uptake" in Figure DR1) is used which again goes through the origin as well as the modern condition, but asymptotes towards a maximum uptake of double modern silicification at infinitely high [H_4SiO_4]:

$$F_{OPAL} = 520 \text{ Tmol Si/yr} * [H_4SiO_4]_{SURF} / ([H_4SiO_4]_{SURF} + .0077)$$

In response to PETM forcing using the "linear uptake" parameterization, surface [H_4SiO_4] approaches a doubling, whereas due to the weaker response of uptake to elevated [H_4SiO_4] in the "asymptotic uptake" case, the surface box approaches ~5 times initial [H_4SiO_4].

In addition, the model was configured to simulate the significantly reduced Si uptake efficiency (relative to modern) that might have resulted from the somewhat reduced prevalence of diatoms during the Paleogene. Diatoms are the most important silicifiers in the modern ocean, but they only originated during the Jurassic and slowly rose to their modern prominence over the Cretaceous and the early part of the Cenozoic

(Harper and Knoll, 1975; Racki and Cordey, 2000), so they could have played a less important role during the PETM. In these runs (denoted "1/10th uptake" in Figure DR1) the linear uptake parameterization was scaled down to 10%:

$$F_{OPAL} = 0.1 * [H_4SiO_4]_{SURF} / .0077 * 260 \text{ Tmol Si/year}$$

These cases represent extreme reductions in Si uptake efficiency, probably far lower than the pre-diatom world, and were designed to constrain the maximum effect that the reduced prevalence of diatoms might have had on Si-cycling during the PETM. All configurations featuring reduced Si uptake efficiency were spun up to a new steady state by running the model for millions of years with constant riverine input. In order to balance that riverine Si input with Si burial (requiring similar total surface opal production to the "linear uptake" at reduced uptake efficiency, the steady state of those configurations features 10 times higher surface $[H_4SiO_4]$ (or slightly different depending on opal burial parameterizations, described below).

Opal export dissolution vs. burial: The standard version of the model (denoted "constant dissolution" in Figure DR1) uses fixed fractions of opal production that remineralize in the surface (50%) and deep ocean (47.42%), with the rest (2.58%) being buried. However, it is likely that an increase (or decrease) in water column $[H_4SiO_4]$ would cause more (or less) exported opal to survive dissolution and be buried in sediments (Sarmiento and Gruber, 2006). To account for this, two schemes for responsive partitioning of opal export between dissolution and burial were employed. In the first (denoted "dissolution sensitivity" in Figure DR1), opal burial (in mol Si/year) is parameterized as a linear function of deep $[H_4SiO_4]$ going through the origin and modern steady-state point of the default model configuration (burial of 6.7 Tmol Si/year at deep $[H_4SiO_4]$ = 122.9 mmol/m^3):

$$F_{BURIAL} = 6.7 \text{ Tmol Si/year} * [H_4SiO_4]_{DEEP} / 0.1229$$

In the second parameterization (denoted “burial fraction sensitivity” in Figure DR1), the fraction of opal production that survives dissolution to be buried in sediments is parameterized as a linear function of deep $[H_4SiO_4]$ going through the origin and the steady-state point of the default model configuration (2.58% of opal production is buried at deep $[H_4SiO_4]$ = 122.9):

$$fr_{BURIAL} = .0258 * [H_4SiO_4]_{DEEP} / 0.1229$$

$$F_{BURIAL} = fr_{BURIAL} * F_{OPAL}$$

In both of these parameterizations of burial rate, the remainder of opal production (that does not dissolve in the surface ocean or get buried in sediments) dissolves in the deep ocean:

$$F_{DISS\text{-}DEEP} = F_{OPAL} - F_{DISS\text{-}SURF} - F_{BURIAL}$$

Each burial scheme can be coupled with each parameterization of opal uptake, as well as a model run in which opal uptake does not respond at all to surface $[H_4SiO_4]$ (denoted “constant uptake” in Figure DR1), leaving the response of opal burial to deep $[H_4SiO_4]$ as the only stabilizing feedback in the model.

References

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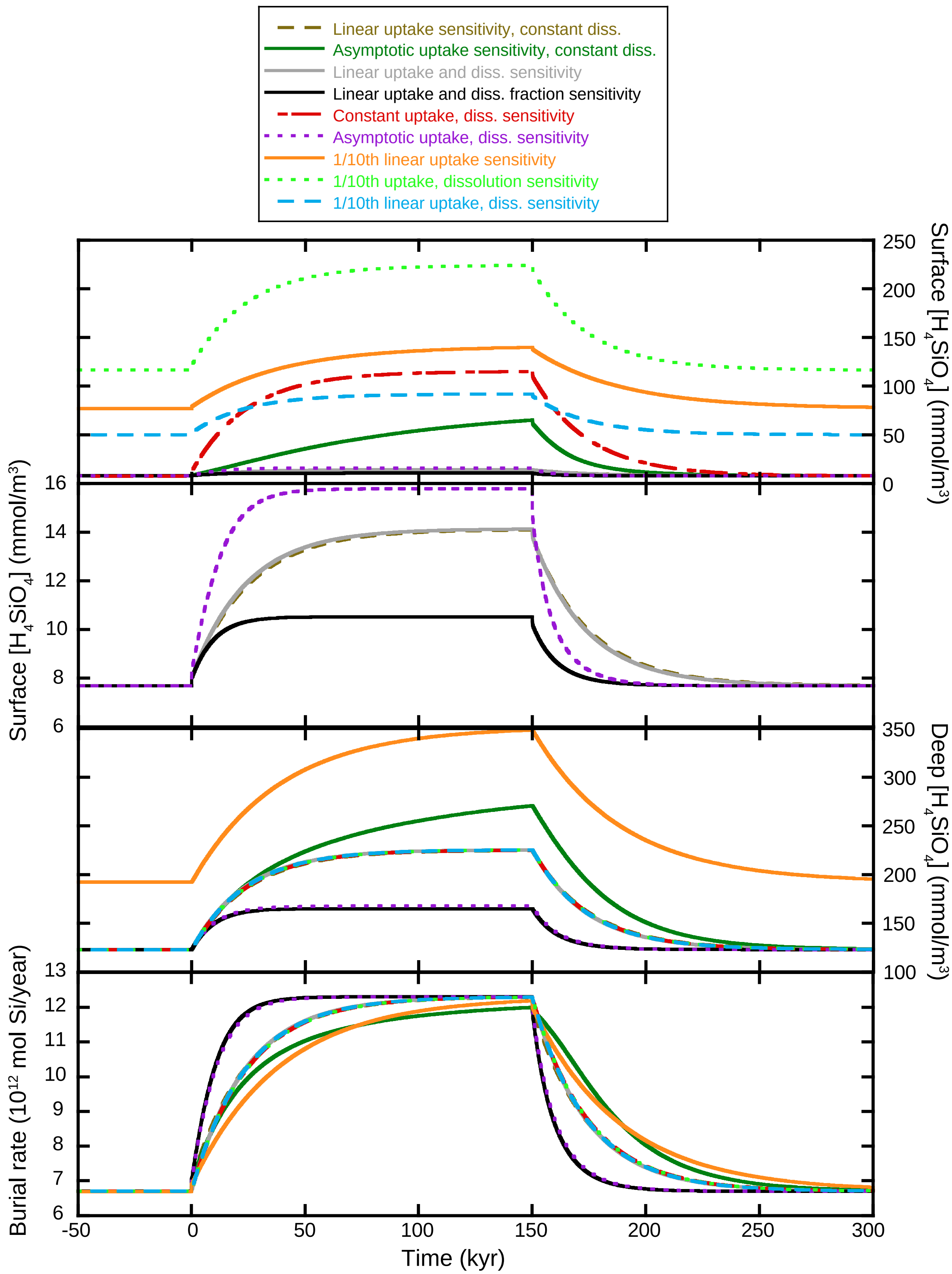
Figure DR1: Modeled surface and deep [H_4SiO_4] and opal burial rate response to elevated riverine Si input during the PETM using various combinations of Si-cycle feedbacks. In all cases riverine flux is doubled (corresponding to the maximum PETM C release estimate) during the interval 0 to 150 kyr. See Supplemental text for full description of response functions.



Linear uptake sensitivity, constant diss.
Asymptotic uptake sensitivity, constant diss.
Linear uptake and diss. sensitivity
Linear uptake and diss. fraction sensitivity
Constant uptake, diss. sensitivity
Asymptotic uptake, diss. sensitivity
1/10th linear uptake sensitivity
1/10th uptake, dissolution sensitivity
1/10th linear uptake, diss. sensitivity
Surface [H4SiO4] (mmol/m3)
Surface [H4SiO4] (mmol/m3)
Deep [H4SiO4] (mmol/m3)
Burial rate (10^12 mol Si/year)
Time (kyr)