

Supplementary Text

We calculated the global mean surface temperature (T_s) using the following equation:

$$T_s = T_{CO_2+H_2O} + \Delta T_{CH_4+C_2H_6}. \quad (10)$$

The surface temperature that reflects the greenhouse effect of CO_2 and H_2O ($T_{CO_2+H_2O}$) was calculated using the climate parameterization created by Lehmer et al. (2020):

$$\begin{aligned} T_{CO_2+H_2O} = & (4.809 - 222.0 X - 68.44 X^2 - 6.737 X^3 - 0.206 X^4 \\ & + 1414 XY + 446.4 X^2 Y + 44.41 X^3 Y + 1.364 X^4 Y \\ & - 2964 XY^2 - 978.4 X^2 Y^2 - 98.86 X^3 Y^2 - 3.059 X^4 Y^2 \\ & + 2655 XY^3 + 907.5 X^2 Y^3 + 92.87 X^3 Y^3 + 2.892 X^4 Y^3 \\ & - 868.4 XY^4 - 304.6 X^2 Y^4 - 31.48 X^3 Y^4 - 0.985 X^4 Y^4 \\ & + 1045 Y - 1496 Y^2 + 1064 Y^3 - 281.1 Y^4), \end{aligned} \quad (11)$$

where $X = \ln(pCO_2)$ and Y is solar luminosity relative to present.

The surface temperature change due to methane and ethane was calculated following Haqq-Misra et al. (2008). We used the calculated surface temperature at $f_{CH_4} = 0, 10^{-5}, 10^{-4}, 10^{-3}, 3 \times 10^{-3}$ and 10^{-2} , without considering the radiative effect of haze (Haqq-Misra et al. 2008) (in their Figure 8). Here, we decided to use the radiative effect of methane and ethane at $pCO_2 = 0.06$ bar, for which the effect of hydrocarbon haze is expected to be small, even at high f_{CH_4} (10^{-2}). This treatment is possible because of the small overlap of wavelengths for the strong absorption of CO_2 ($\sim 15 \mu m$), CH_4 ($\sim 8 \mu m$), and C_2H_6 ($\sim 12 \mu m$) (Byrne and Goldblatt 2014a, b). The pCO_2 dependence on $\Delta T_{CH_4+C_2H_6}$ is minor for the range where hydrocarbon haze is not formed ($CH_4/CO_2 \ll 0.2$) (Haqq-Misra et al. 2008). The value of $\Delta T_{CH_4+C_2H_6}$ was interpolated linearly (log-linearly) when pCH_4 was smaller (larger) than 10^{-5} bar using the following equation:

$$\Delta T_{CH_4+C_2H_6} = a_{CH_4} \cdot (pCH_4)^{b_{CH_4}}. \quad (12)$$

The values of a_{CH_4} and b_{CH_4} are listed in Supplementary Table 2. When pCH_4 was larger than 10^{-2} bar, the dependency was extrapolated using the linear relationship between $pCH_4 = 3 \times 10^{-3}$ bar and 10^{-2} bar.

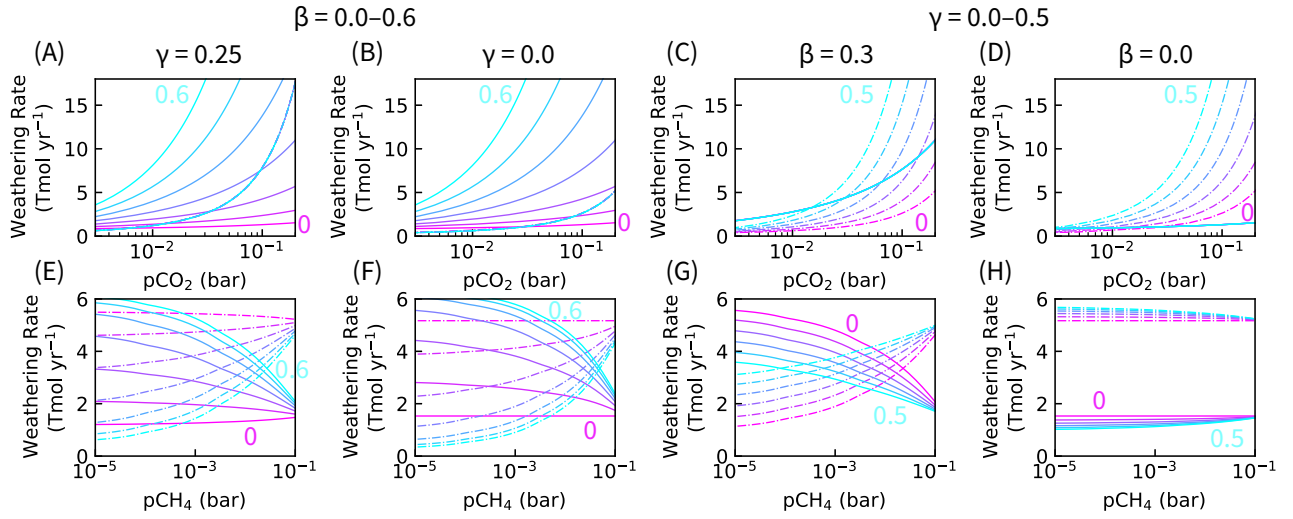
The deep-sea temperature (T_d) and pore-space temperature (T_{pore}) were related to the global mean surface temperature (T_s) according to Krissansen-Totton et al. (2018):

$$T_d = 1.1 \cdot T_s - 39.463 \text{ (K)}. \quad (13)$$

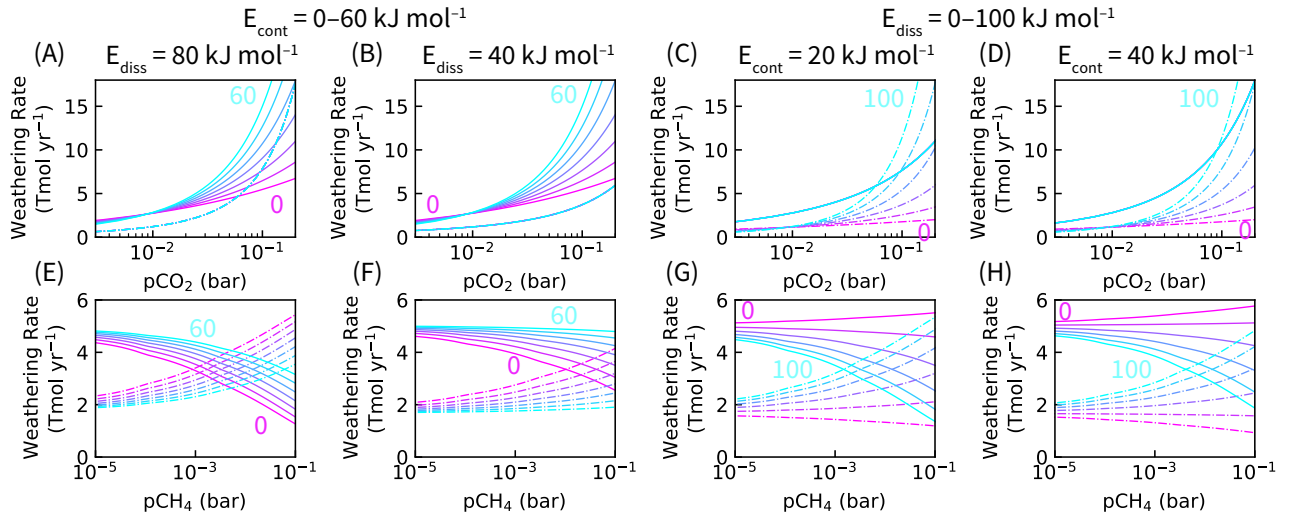
$$T_{pore} = T_d + 9.0. \quad (14)$$

References

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Supplementary Figures

Supplementary Figure 1. Response of continental/seafloor weathering rate (Tmol yr^{-1}) (solid line/dot-dashed line) to (A–D) CO_2 -induced warming at fixed $p\text{CH}_4$ (10^{-4} bar) and to (E–H) CH_4 -induced warming at the present CO_2 outgassing rate (6.7 Tmol yr^{-1}) with different sets of the parameter that represents the CO_2 -dependence of continental weathering (β) and the $p\text{H}$ -dependence of seafloor weathering (γ). Different colors represent different values of β (0.0, 0.1, 0.2, 0.3, 0.4, 0.5, and 0.6) (A–B, E–F) or γ (0.0, 0.1, 0.2, 0.3, 0.4, and 0.5) (C–D, G–H). Pink (cyan) color represents smaller (larger) values of each parameter. These calculations are conducted with solar luminosity (83% of present Earth), seafloor spreading rate (1.307 times the present rate), soil biological activity ($f_e = 0.15$), and present continental area ($f_a = 1$).



Supplementary Figure 2. Response of continental/seafloor weathering rate (Tmol yr^{-1}) (solid line/dot-dashed line) to (A-D) CO_2 -induced warming at fixed $p\text{CH}_4$ (10^{-4} bar) and to (E-H) CH_4 -induced warming at the present CO_2 outgassing rate (6.7 Tmol yr^{-1}) with different sets of effective activation energy of continental (E_{cont}) and seafloor weathering (E_{diss}). Different colors represent different values of E_{cont} (0, 20, 40, 60, and 80 kJ mol^{-1}) (A-B, E-F) or E_{diss} (0, 20, 40, 60, 80, and 100 kJ mol^{-1}) (C-D, G-H). Pink (cyan) color represents smaller (larger) values of each parameter. These calculations are conducted with solar luminosity (83% of present Earth), seafloor spreading rate (1.307 times the present rate), soil biological activity ($f_e = 0.15$), and present continental area ($f_a = 1$).

Supplementary Tables

Parameter	Value	Remarks and References
$T_{s,0}$	285 (K)	Krissansen-Totton et al. (2018)
$pCO_{2,0}$	280 (ppm)	Krissansen-Totton et al. (2018) *For the unit conversion to PAL, 300 ppm is used in this study.
$[H^+]_{pore,0}$	1.1223×10^{-9} (mol L ⁻¹)	Corresponds to $pCO_{2,0}$ using Equation (4)
$F_{ws,0}$	6.25 (Tmol yr ⁻¹)	Berner (1991), Krissansen-Totton et al. (2018)
$F_{wc,0}$	13.3 (Tmol yr ⁻¹)	Berner (1991)
$F_{sws,0}$	0.45 (Tmol yr ⁻¹)	Krissansen-Totton et al. (2018)
$F_{rp,0}$	0.09 (Tmol yr ⁻¹)	Harada et al. (2015)

Supplementary Table 1. List of the parameters that represent the condition of the modern Earth.

Range of $p\text{CH}_4$ (bar)	a_{CH_4}	b_{CH_4}
$p\text{CH}_4 < 10^{-5}$	201264.931	1.0
$10^{-5} \leq p\text{CH}_4 < 10^{-4}$	400.896	0.4598527
$10^{-4} \leq p\text{CH}_4 < 10^{-3}$	68.912	0.2686689
$10^{-3} \leq p\text{CH}_4 < 3 \times 10^{-3}$	66.072	0.2625762
$3 \times 10^{-3} \leq p\text{CH}_4 < 10^{-2}$	43.475	0.1905246
$10^{-2} \leq p\text{CH}_4$ (extrapolated)	43.475	0.1905246

Supplementary Table 2. The values of parameters used for calculating the greenhouse effect of CH_4 and C_2H_6 .