S1. Model options

S1.1. Signal tracking method

S1.1.1. Time-stepping method

IMP can adopt the time-stepping method (method 1 in Fig. 2a) as an option. In this section, the time-stepping method is used for dissolution experiment #2 of Section 3.2.2 and results are 5 compared with those by method 2. The isotope signal changes in δ^{13} C and δ^{18} O are discretized into 42 steps, represented by 42 classes of $CaCO_3$ particles with distinct signals (Fig. S1) as described in Section 2.5.1.

When using the time-stepping method with 42 different CaCO₃ classes for dissolution experiment #2, very similar results are obtained to those with the interpolating method (com-10 pare Figs. S2a–c and S2d–f). Results of the time-stepping method approach the interpolation method results when the total number of $CaCO_3$ classes is increased. Furthermore, other simulations adopting the time-stepping method lead to the same degree of agreement of the results (e.g., dissolution experiment #1). Such comparisons confirm that signal tracking diagenesis can be simulated by either method 1 or 2 but a large number of $CaCO_3$ classes is required when 15

adopting method 1.

S1.1.1. Direct tracking of CaCO₃ isotopologues

As another option, IMP is able to directly track temporal changes of spatial distributions of five $CaCO_3 \ isotopologues: \ Ca^{12}C^{16}O_3, \ Ca^{12}C^{18}O^{16}O_2, \ Ca^{13}C^{16}O_3, \ Ca^{13}C^{18}O^{16}O_2 \ and \ Ca^{14}CO_3.$ This option is a specific application of the direct tracking method (method 3 in Fig. 2c) and is beneficial because it allows tracking of ¹⁴C age and Δ_{47} , in addition to δ^{13} C and δ^{18} O, in a computationally efficient way (Section 2.5.1). For ${}^{14}C$ age tracking the radioactive decay of $Ca^{14}CO_3$ and accompanying generation of alkalinity need be accounted for:

$$R_{\ell} = (1-\phi)m_{\ell}k_{\rm cc,\ell}(1-\Omega_{\rm cc})^{\eta_{\rm cc}}H(1-\Omega_{\rm cc}) + (1-\phi)m_{\ell}\lambda_{\ell}$$
(S1)

$$R_{\rm ALK} = (1-\phi)m_{\rm OM}k_{\rm anox} + \sum_{\ell=1}^{n_{\rm cc}} 2(1-\phi)m_{\ell}k_{\rm cc,\ell}(1-\Omega_{\rm cc})^{\eta_{\rm cc}}H(1-\Omega_{\rm cc}) + (1-\phi)m_{\ell}\lambda_{\ell} \quad (S2)$$

where ℓ is one of $n_{\rm cc}$ CaCO₃ classes including Ca¹⁴CO₃ and Ca¹²CO₃, and λ_{ℓ} is the decay constant which is 8033^{-1} yr⁻¹ for Ca¹⁴CO₃ (Aloisi et al., 2004) and zero for all other classes. With method 3, Eqs. (S1) and (S2) can be implemented instead of Eqs. (5) and (9) for ${}^{14}C$ age tracking. Apart from the implementation of Eqs. (S1) and (S2) instead of Eqs. (5) and (9) the model calculation and application to signal tracking are the same as described in Section 2.

As an example for the direct tracking of $CaCO_3$ isotopologues, we conduct again dissolution experiment #2 as presented in Section 3.2.2. Here changes in Δ_{47} in CaCO₃ are imposed additionally and are assumed synchronous with the δ^{18} O changes (Fig. S3). The rain fluxes of 25 individual isotopologues (Fig. S3b) are calculated directly from the input signals (Fig. S3a),

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except for the $Ca^{14}CO_3$ flux, which is calculated assuming that the ${}^{14}C/{}^{12}C$ ratio is constant at 1.2×10^{-12} (Aloisi et al., 2004). Note that the changes in rain fluxes with time are hard to discern from the semi-log plot of Fig. S3b. Another set of the five isotopologues needs to be additionally simulated when one further tracks model time as a proxy.

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Obtained signals of δ^{13} C and δ^{18} O as well as CaCO₃ abundance (Fig. S4a–c) are the same as those obtained by method 2 (Figs. 12g-i), thus validating the direct tracking method. As the imposed temporal changes for Δ_{47} are the same as those for δ^{18} O (Fig. S3a), the same patterns of signal distortion are obtained for both signals (Figs. S4b and e). The ¹⁴C age is increased from ~ 5 up to ~ 50 kyr by dissolution (Fig. S4d), which is consistent with previous

results (e.g., Keir, 1984; Broecker et al., 1991; Oxburgh, 1998; Barker et al., 2007), but with our model, we can further examine the effects of different styles of bioturbation as well as different extents of dissolution (e.g., Fig. S4d). Note, however, that ¹⁴C ages plotted against diagnosed depth do not reflect ${}^{14}C$ decay below the mixed layer (Section 2.5.2 and Fig. 4).

As an interesting feature of the option, we can further examine the kinetic isotope effect 40 on signal distortion. As an example, assuming -0.005% of kinetic isotope effect for dissolution of Ca¹³C¹⁸O¹⁶O₂ (i.e., $k_{\rm cc,Ca^{13}C^{18}O^{16}O_2} = (1 - 5 \times 10^{-5}) \times 365.25 \ {\rm yr}^{-1}$), quite large signal distortions on Δ_{47} are predicted (dotted curves in Fig. S4f). Although the kinetic isotope effect for CaCO₃ dissolution is not known due to the lack of experimental report (DePaolo, 2011), we can examine a hypothetical kinetic isotope effect for any isotopologues (not limited 45

to $Ca^{13}C^{18}O^{16}O_2$) on signal distortion by changing $k_{cc,\ell}$ values. For example, simulations with various extents of dissolution (e.g., dissolution experiment #1 of Section 3.2.2) and $k_{cc,\ell}$ values suggest that signal distortion can be dominated by the kinetic isotope effect when the kinetic isotope effect is relatively large and/or dissolution is intense.

S1.2. Utilization of CO_2 chemistry calculation modules 50

IMP has the option to adopt improved routines to model the ocean carbonate system, i.e., mocsy (Orr and Epitalon, 2015, for the Fortran and Python versions) and CO2SYS (Lewis and Wallace, 1998; van Heuven et al., 1998; Humphreys et al., 2020, for the Python and MATLAB versions), as a replacement for the simplified calculation summarized in Tables 2 and 3. With the more complicated and realistic calculations of CO₂ chemistry by mocsy and CO2SYS, diagenetic 55 results are modified, but responses of CaCO₃ to undersaturation are not so significantly affected (Figs. S5 and S6, compare with Figs. 7 and 8). Nonetheless, the option is beneficial because changes in CO_2 chemistry caused by a comprehensive set of environmental factors (including aqueous species other than carbonate species) can be explicitly reflected in CaCO₃ diagenesis and proxy signals.

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S1.3. Time tracking

We illustrated signal tracking diagenesis with the interpolating method (method 2, Section 2.5.1) in Section 3.2. Simulations in Sections 3.2.1 and 3.2.2 track 2 paleoceanographic proxies δ^{13} C and δ^{18} O with 4 classes of CaCO₃ particles that have the maximum and/or minimum values of

input δ¹³C and δ¹⁸O. In Section 3.2.3, we doubled the number of CaCO₃ classes (i.e., adopted 8 classes) to track the δ¹³C and δ¹⁸O in two distinctive model species ('fine' vs. 'coarse' species). By adopting 8 classes of CaCO₃ particles that have the endmember values of δ¹³C and δ¹⁸O and size-dependent dissolution and mixing properties, one can track not only δ¹³C and δ¹⁸O, but also the size distribution in bulk CaCO₃ in simulations in Section 3.2.3 (cf. Fig. 14c). Thus, a
simple relationship can apply to method 2: when one tracks n_p proxies and/or characteristics (e.g., size) of bulk CaCO₃, 2^{n_p} classes of CaCO₃ particles are necessary as described in Section

2.5.1.

Trackable characteristics with method 2 include model time (Section 2.5.2). As an example, consider a simplest case where one tracks only the average model time in bulk CaCO₃ but not ⁷⁵ any other proxies or characteristics. This case requires two classes of CaCO₃ particles with the minimum (or start) and maximum (or final) model-time. The model time can be tracked by assigning rain fluxes to the two classes of CaCO₃ particles so that flux-weighted average mode-time of the two classes matches the model time of the simulation. For example, the total carbonate rain flux is dominated by the CaCO₃ class with the minimum model-time at the start of the simulation, equally contributed by the two classes at the middle of the simulation and is dominated by the class with the maximum model-time at the end of simulation. More generally, one can additionally track model time by doubling the number of CaCO₃ classes that are used for simulations without time tracking.

As examples, the following subsections (Sections S1.3.1–S1.3.3) repeat the same simulations as in Sections 3.2.1–3.2.3 but with enabling tracking of model time by adopting 8 or 16 classes of CaCO₃ particles whose properties are tabulated in Tables S1 and S2. Except for the doubled numbers of simulated CaCO₃ classes (Tables S1 and S2), the experimental conditions are the same as those presented in Section 3.2.

S1.3.1. Bioturbation

⁹⁰ The experiments to examine distortions of proxy signals by bioturbation in Section 3.2.1 are repeated in this section with additionally tracking model time (Fig. S7d) by simulating 8 classes of CaCO₃ particles in Table S1.

Before describing the results, the details on the calculation of the rain fluxes for individual CaCO₃ classes are presented first. When one considers a given proxy or characteristic, CaCO₃ classes can be grouped into 2 sets of classes, which have either the maximum or minimum value for the cosidered proxy/characteristic (Table S1). The rain fractions for the two sets can be calculated from the input value for the proxy/characteristic (e.g., Fig. S7b when considering

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 δ^{13} C). For instance, if the two sets of CaCO₃ classes have the values of A and B and the input value is given as X (cf. Fig. 2b), then the rain fractions for the two sets of classes (denoted as a and b, respectively) can be calculated as a solution of two equations: aA + bB = X and a + b = 1. After repeating the assignment of the rain fractions for two sets of classes for all the considered proxies/characteristics (Figs. S7a–d), the rain fraction of a given class can be calculated as the product of the rain fractions for the sets that have the same properties as those of the considered class (Fig. S7e). As an example, rain fraction of class #1 (dotted black curve in Fig. S7e) is calculated as the product of the rain fractions for the sets of classes that have the maximum values for δ^{13} C, δ^{18} O and model time (solid curves in Figs. S7b–d, respectively).

The age model (the relationship between model time and diagnosed depth) is an additional output of the model by enabling time tracking (Figs. S8c and g). The age model is the same among different cases with and without bioturbation when disabling dissolution because sediment accumulates at the same rate below the mixed layer (Fig. S8c). Thus, with dissolution disabled, depth-dependent proxy signal changes can be directly converted to temporal changes with a single age model (compare Figs. S8a and b with Figs. S9a and b).

On the other hand, a single age model cannot be applied to reveal temporal changes of proxy signals when carbonate dissolution is mechanistically linked to styles of bio-mixing (Figs. S8g and h). Apparent signal shifts in depth caused by changes in the extent of dissolution or sediment accumulation rate (Figs. S8e and f) are absent when plotted against mode time using individual age models (Figs. S9d and e). It also becomes clearer that proxy signals are more effectively lost along with CaCO₃ particles when mixing is relatively ineffective. For example, slight reductions of δ^{18} O peaks by enabling dissolution are recognized only for cases with no bioturbation and LABS mixing (see slightly smaller peaks of pink and orange curves in Fig. S9e than in Fig. S9b). Note that LABS mixing may cause a net upward transport and thus be less efficient at deep depths than homogeneous and Fickian mixing (Section 3.2.1, Fig. S8b).

S1.3.2. Dissolution of carbonates

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The same dissolution experiments as in Section 3.2.2 (Fig. 11b) are repeated in this section except with enabling model-time tracking. Instead of 4 we adopt 8 classes (Table S1) to track model time in addition to δ^{13} C and δ^{18} O. The temporal changes of the rain fractions for the 8 CaCO₃ classes are presented in Fig. S10b, calculated in a way described in Section S1.3.1 (Table S1 and Fig. S10b).

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Perfect age models for different cases with and without bioturbation are available with enabling time tracking, with which signal change events recorded in the sediment (i.e., Fig. 12) can be plotted against model time (Fig. S11). Signal plots against model time reveal that long-term input signals are reconstructed reasonably well when dissolution is not so severe (as in experiment #1, Figs. S11a-f). However, when signals are lost by intense dissolution as in experiment #2, signal change events are significantly obscured (Figs. S11g-i). Preservation of

proxy signals is better with efficient bioturbation (e.g., non-local homogeneous mixing; green 135 curves in Figs. S11g and h) because bio-mixing can transfer CaCO₃ particles with preserved signals to locations where proxy signals are lost by dissolution along with $CaCO_3$ particles.

S1.3.3. Species-specific mixing/dissolution

Differences in recorded proxy signals in two distinctive $CaCO_3$ species with different grain sizes ('coarse' vs. 'fine' speces) are examined by simulating 8 classes of $CaCO_3$ particles in Section 140 3.2.3. Here we repeat the same experiments but with additionally tracking model time by simulating 16 classes of $CaCO_3$ particles in Table S2 (Fig. S12).

Using the tracked time against diagnosed depth (i.e., age model) proxy signals in the two distinctive $CaCO_3$ species can be plotted against model time (Fig. S13). As described in Section 3.2.3, apparent offsets of excursions of proxy signals and species abundance between the two 145 species disappear (compare Figs. S13 and 14) but the reduction of the magnitude of imposed species abundance changes is still recognized especially in cases with bioturbation (Fig. S13c).

References

- Aloisi, G., Wallmann, K., Haese, R. R., and Saliège, J. F. (2004). Chemical, biological and hydrological controls on the ¹⁴C content of cold seep carbonate crust: numerical modeling 150 and implications for convection at cold seeps. Chemical Geology, 213:359–383.
 - Barker, S., Broecker, W., Clark, E., and Hajdas, I. (2007). Radiocarbon age offsets of for a for a form differential dissolution and fragmentation within the sedimentary bioturbated zone. Paleoceanography, 22(2).
- Broecker, W. S., Klas, M., Clark, E., Bonani, G., Ivy, S., and Wolfli, W. (1991). The influence 155 of $CaCO_3$ dissolution on core top radiocarbon ages for deep-sea sediments. *Paleoceanography*, 6(5):593-608.
 - DePaolo, D. J. (2011). Surface kinetic model for isotopic and trace element fractionation during precipitation of calcite from aqueous solutions. Geochimica et Cosmochimica Acta, 75:1039-1056.

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- Humphreys, M. P., Gregor, L., Pierrot, D., van Heuven, S. M. A. C., Lewis, E. R., and Wallace, D. W. R. (2020). PyCO2SYS: marine carbonate system calculations in Python. Zenodo.
- Keir, R. S. (1984). Recent increase in Pacific CaCO₃ dissolution: A mechanism for generating old ¹⁴C ages. Marine Geology, 59(1-4):227-250.
- Lewis, E. and Wallace, D. W. R. (1998). Program Developed for CO₂ System Calculations. 165 ORNL/CDIAC-105.

- Orr, J. C. and Epitalon, J. M. (2015). Improved routines to model the ocean carbonate system: mocsy 2.0. *Geoscientific Model Development*, 8:485–499.
- Oxburgh, R. (1998). The Holocene preservation history of equatorial Pacific sediments. *Paleoceanography*, 13(1):50–62.
 - van Heuven, S., Pierrot, D., Rae, J., Lewis, E., and Wallace, D. (1998). MATLAB Program Developed for CO₂ System Calculations. *ORNL/CDIAC-105b*.

Property ^a	$CaCO_3$ class #										
	1	2	3	4	5	6	7	8			
$\delta^{13} \mathrm{C}$	+	+	_	_	+	+	_	_			
$\delta^{18} \mathrm{O}$	+	_	+	_	+	_	+	_			
Time	+	+	+	+	_	_	_	_			

Table S1: Properties of CaCO₃ classes in simulations in Sections S1.3.1 and S1.3.2.

^a '+' and '-' denote the maximum and minimum values for CaCO₃ property. As an example, when δ^{18} O is tracked and its input values are between -1 and 1 ‰ for a given simulation, CaCO₃ classes with '-' and '+' for δ^{18} O have -1 and 1 ‰ of δ^{18} O, respectively.

Table S2: Properties of $CaCO_3$ classes in simulations in Section S1.3.3.

Property ^a	$CaCO_3$ class #															
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
$\delta^{13}C$	+	+	_	_	+	+	_	_	+	+	_	_	+	+	_	_
$\delta^{18}{ m O}$	+	_	+	_	+	_	+	_	+	_	+	_	+	_	+	_
Size	\mathbf{F}	\mathbf{F}	\mathbf{F}	\mathbf{F}	\mathbf{C}	\mathbf{C}	\mathbf{C}	\mathbf{C}	F	F	F	F	С	С	С	С
Time	+	+	+	+	+	+	+	+	_	_	_	_	_	_	_	_

^a '+' and '-' denote the maximum and minimum values for CaCO₃ property. As an example, when δ^{18} O is tracked and its input values are between -1 and 1 % for a given simulation, CaCO₃ classes with '-' and '+' for δ^{18} O have -1 and 1 % of δ^{18} O, respectively. 'F' and 'C' denote that the grain size of a CaCO₃ classe is 'fine' and 'coarse', respectively. 'Coarse'-sized classes have the standard mixing and dissolution properties while classes of 'fine' size are mixed to a deeper depth (20 cm) and dissolved faster (10 times the standard rate).



Figure S1: Timelines of proxy inputs (a), rain fluxes of individual classes of $CaCO_3$ particles (b) with different proxy values (d) and water depth changes (c) in simulations illustrating the option to adopt the time-stepping method. Except for the number of $CaCO_3$ classes and assignment of proxy and flux values to individual classes (b and d), the simulation conditions are the same as those in dissolution experiment #2 in Fig. 11.



Figure S2: Proxy signals (a, b, d and e) and weight fraction of bulk CaCO₃ in solid sediment (c and f) in dissolution experiment #2 in Fig. 11, tracked by the time-stepping method (method 1) with 42 classes of CaCO₃ particles (a–c) and by the interpolating method (method 2) with 4 classes of CaCO₃ particles (d–f).



Figure S3: Timelines of proxy inputs (a), rain fluxes of individual CaCO₃ isotopologues (b) and water depth changes (c) in simulations illustrating the option of direct tracking of CaCO₃ isotopologues. In a, δ^{13} C and δ^{18} O are plotted against left vertical axis while Δ_{47} is plotted against right vertical axis. The total rain flux of CaCO₃ and time changes of δ^{13} C, δ^{18} O and water depth are the same as those in dissolution experiment #2 in Fig. 11. Note that in b time changes in rain fluxes of individual CaCO₃ isotopologues are too small to be discerned on the logarithmic scale.



Figure S4: Proxy signals (a, b, d, e and f) and weight fraction of bulk CaCO₃ in solid sediment (c) tracked by 5 CaCO₃ isotopologues in simulations illustrating the option to directly track CaCO₃ isotopologues. In f, Δ_{47} distortion caused by diagenesis including a hypothetical kinetic isotope effect (KIE) of -0.005% for Ca¹³C¹⁸O¹⁶O₂ dissolution is depicted by dotted curves.



Figure S5: Estimated CaCO₃ weight fractions in mixed layer and burial fluxes as functions of CaCO₃ saturation degree (Δ CO₃) and rain fluxes, with enabling only oxic degradation of organic matter. The results shown are from the model which adopts a shallower sediment depth (50 cm), single class of CaCO₃ particles, Fickian mixing for bioturbation and mocsy to calculate CO₂ chemistry.



Figure S6: As for Fig. S5 except enabling both oxic and anoxic degradation of organic matter.



Figure S7: Timelines of proxy inputs (a) and rain fractions for paris of $CaCO_3$ classes that have the maximum and minimum proxy values (b and c) and model time (d), and for individual $CaCO_3$ classes (e), in simulations examining signal distortion by bioturbation. See Table S1 for the properties of individual $CaCO_3$ classes.



Figure S8: Proxy signals (a, b, e and f), model time (c and g) and weight fraction of bulk $CaCO_3$ in solid sediment (d and h) tracked by 8 classes of $CaCO_3$ particles plotted against diagnosed depth in simulations examining signal distortion by bioturbation. In a-d, dissolution rate constants of all $CaCO_3$ classes are fixed at zero, while in e-h, at the default value (Table 1).



Figure S9: Proxy signals (a, b, d and e) and weight fraction of bulk $CaCO_3$ in solid sediment (c and f) plotted against model time in simulations examining signal distortion by bioturbation. In a-c, dissolution rate constants of all $CaCO_3$ classes are fixed at zero, while in d-f, at the default value (Table 1).



Figure S10: Timelines of proxy inputs (a), rain fractions of individual classes of $CaCO_3$ particles (b) and water depth changes (c) in simulations examining signal distortion by $CaCO_3$ dissolution. Two different water depth changes are considered, denoted as dissolution experiments #1 and 2 (c). One set of experiments was conducted without changing the water depth for comparison (dotted line in c). See Table S1 for the properties of individual $CaCO_3$ classes.



Figure S11: Proxy signals (a, b, d, e, g and h) and weight fraction of bulk CaCO₃ in solid sediment (c, f and i) tracked by 8 classes of CaCO₃ particles plotted against model time in simulations examining signal distortion by CaCO₃ dissolution. Two different water depth changes are considered, denoted as dissolution experiments #1 and 2, and compared to the case without water depth change, denoted as control. See Fig. S10c for the assumed water depth changes.



Figure S12: Timelines of proxy inputs (a) and rain fractions of fine and coarse $CaCO_3$ species (b) and individual classes of $CaCO_3$ particles (c) in simulations examining effect of speciesspecific mixing/dissolution properties. See the legend located at the bottom of diagram for the correspondence between $CaCO_3$ classes and types of curve in c. See Table S2 for the properties of individual $CaCO_3$ classes.



Figure S13: Proxy signals (a and b) and weight fraction of bulk $CaCO_3$ in solid sediment (c) for fine and coarse $CaCO_3$ species (solid and dotted curves, respectively) tracked by 16 classes of $CaCO_3$ particles plotted against model time in simulations examining effect of species-specific mixing/dissolution properties.