

Figure A1: Three-isotope plot ($\delta^{30}\text{Si}$ vs $\delta^{29}\text{Si}$) of radiolarian tests (red squares) and sponge spicules (blue squares). The data fall on the expected mass-dependent fractionation (black line: $\delta^{30}\text{Si} = 1.93 \times \delta^{29}\text{Si}$, $r^2=0.998$, $n=105$). Vertical and horizontal bars represent long-term precision (expressed as 2σ).

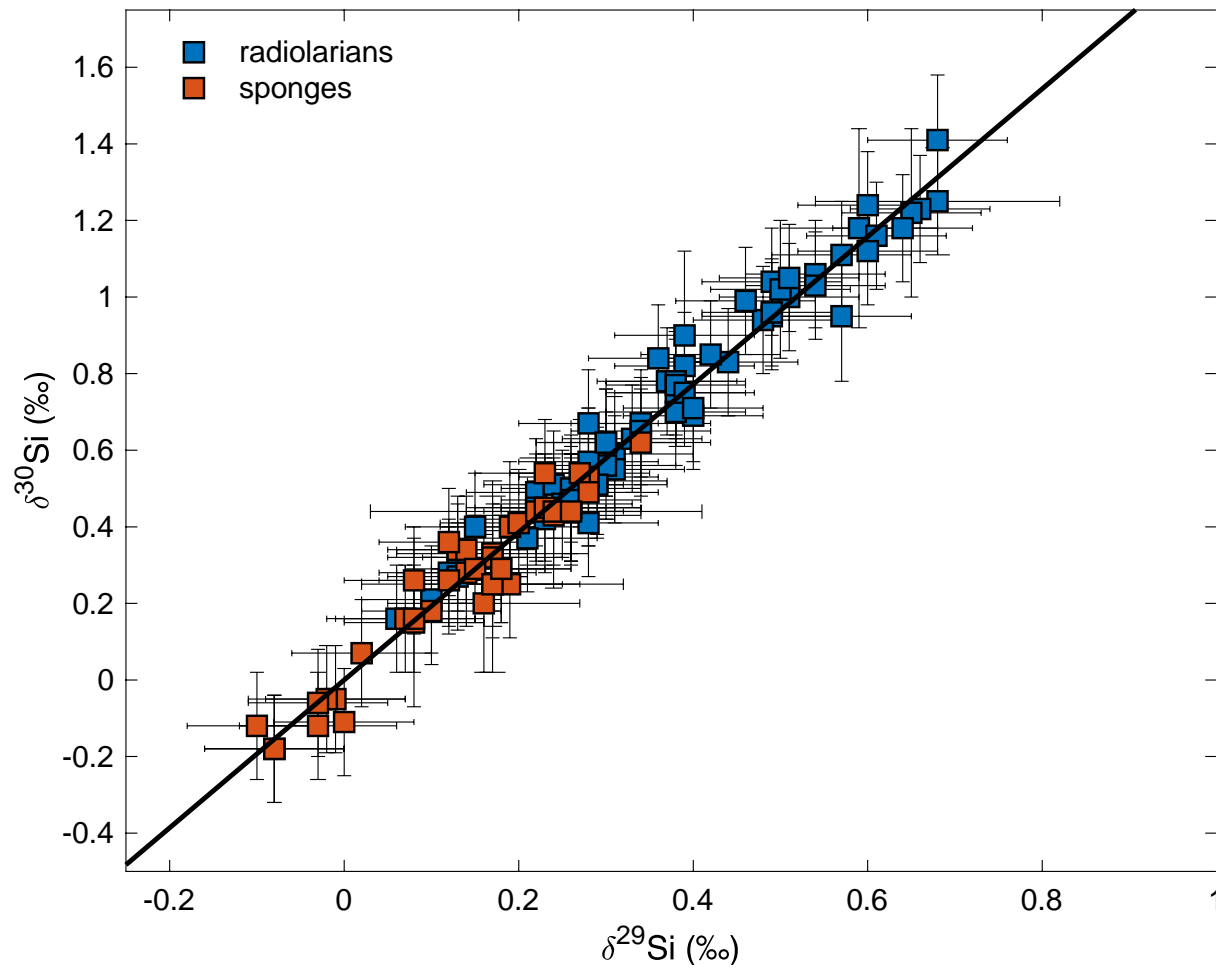


Figure A2: Full dataset of radiolarian tests (blue squares) and sponge spicules (red squares: MC-ICP-MS; red circles: SIMS) $\delta^{30}\text{Si}$. Error bars represent 2σ .

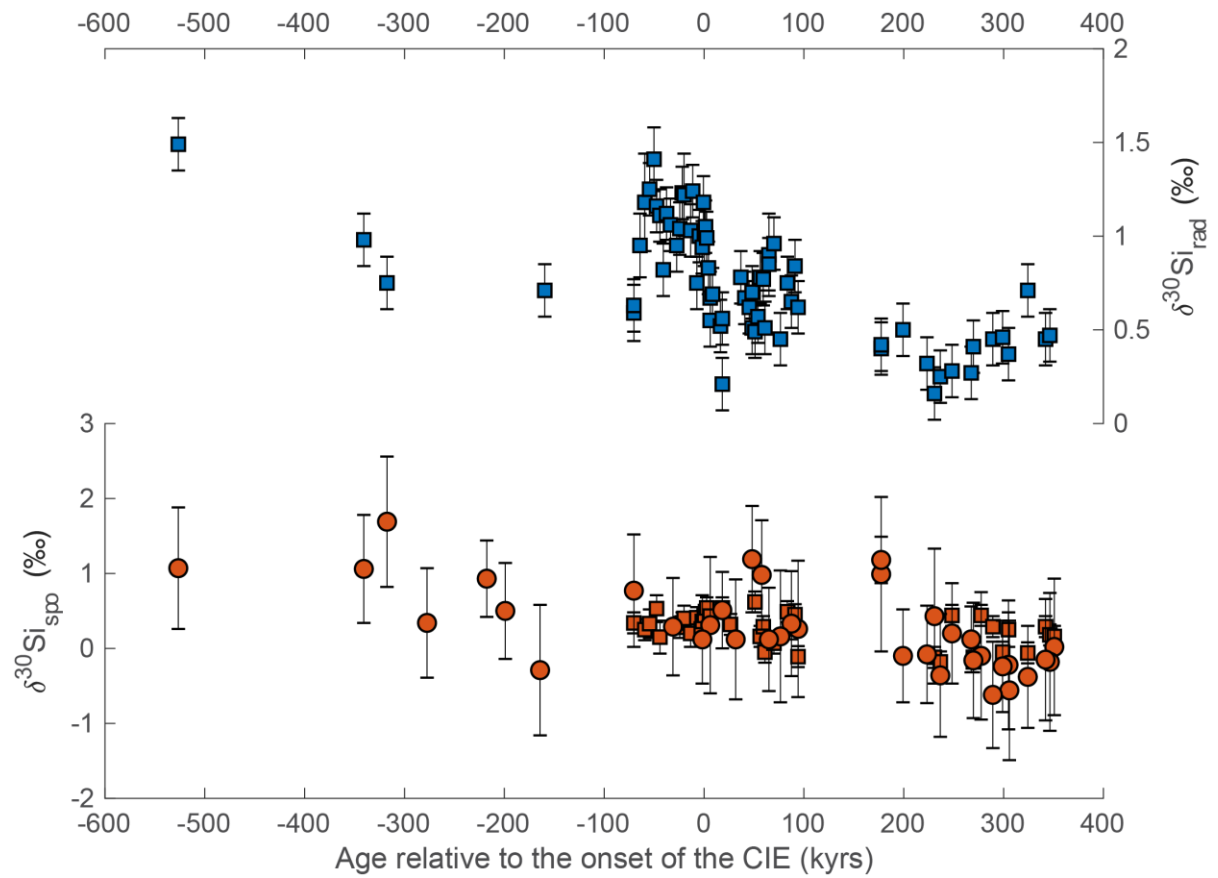


Figure A3: Full dataset of sponge spicules analysis via SIMS. Grey dots represent individual measurements. Red circles show the mean as presented in other figures and data table. Error bars represent 2σ .

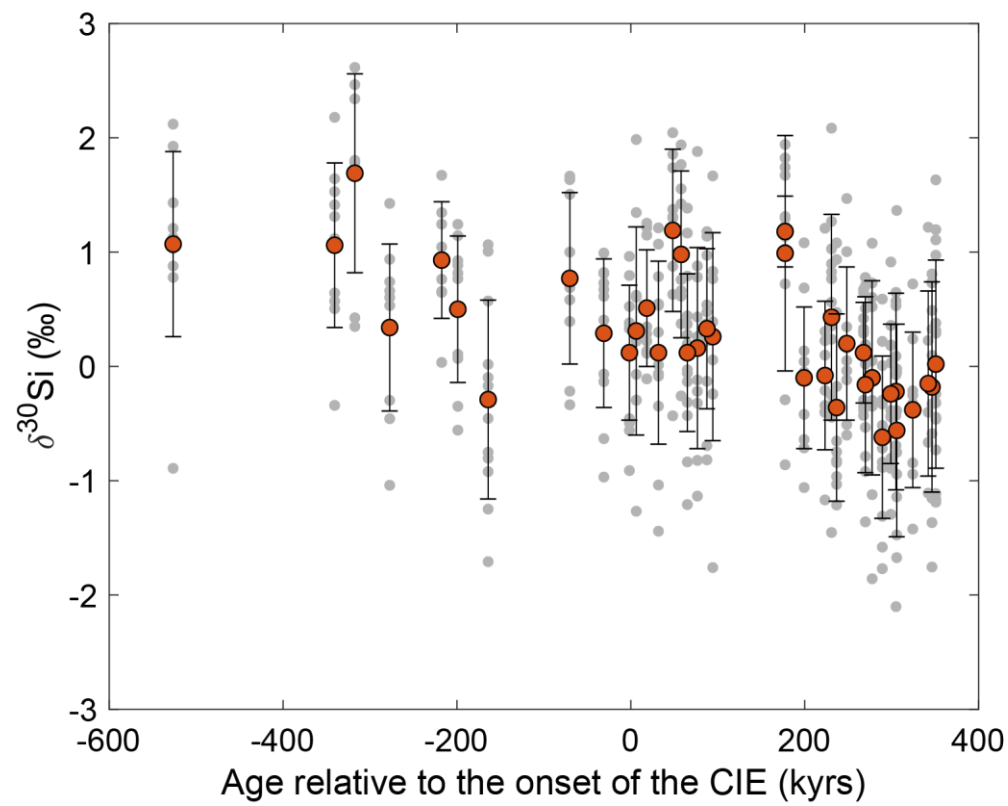


Table A1: Parameters used in the two-box model.

Input	Flux (Tmol/y)	$\delta^{30}\text{Si}$ (‰)	Comment	Reference
Riverine	6.33	1.25	Input from rivers to surface layer	Frings et al. (2016)
Hydrothermal	0.6	-0.3	Input from hydrothermal source to deep layer	Frings et al. (2016)
Dust	0.3	-0.65	Input of dust to surface layer	Frings et al. (2016)
SPM	1.9	-0.18	Input of SPM to surface layer	Frings et al. (2016)
SGD	0.65	0.19	Input of groundwater to surface layer	Frings et al. (2016)
Constant	Value		Comment	Reference
V_{surface}	3.55×10^{19} l		Volume of surface layer	De La Rocha and Bickle (2005)
V_{deep}	1.31×10^{21} l		Volume of deep layer	De La Rocha and Bickle (2005)
V_{exc}	1.37×10^{18} l/y		Yearly water exchange between surface and deep layers	De La Rocha and Bickle (2005)
D_{surf}	98.625 m		Depth of surface layer	
D_{deep}	3651.4 m		Depth of deep layer	
V_{surf}	1800 m/y		Velocity of sinking particles in the surface layer	Passow et al. (2011)
V_{deep}	73000 m/y		Velocity of sinking particles in the deep layer	Passow et al. (2011)
k_{surf}	9		Dissolution constant in the surface layer	Frings et al. (2016)
k_{deep}	27.8		Dissolution constant in the deep layer	Frings et al. (2016)
$[\text{dSi}]_{\text{eq}}$	350×10^{-6} mol/l		Apparent solubility of bSi	Loucaides et al. (2012)
R_{NBS}	0.033532		$^{30}\text{Si}/^{28}\text{Si}$ of standard NBS28	Ding et al. (2005)
α_{diatom}	0.9975		Fractionation associated with diatom production	De La Rocha et al. (1997)
$V_{\text{max_surface}}$	5×10^{14} mol/y		Maximum rate of bSi production in the surface layer	Sarmiento and Gruber (2006)
$V_{\text{max_deep}}$	1.56×10^{12} mol/y		Maximum rate of bSi production in the deep layer	
$K_{\text{M_surface}}$	1×10^{-5} mol/l		Half-saturation constant in the surface layer	Nelson et al. (1995)
$K_{\text{M_deep}}$	1.5×10^{-4} mol/l		Half-saturation constant in the deep layer	

References:

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Table A2: Details of individual samples from hole 1051B and secondary reference material. Uncertainties on $\delta^x\text{Si}$ values are given as individual measurement precision (1σ) for analysis on MC-ICP-MS, and as 2σ for analysis on SIMS. Sample IDs are given as Core-Section-TopInterval.

Sample	Depth (mcd)	Age relative to the PETM (kyrs)	MC-ICP_MS				SIMS			n
			$\delta^{30}\text{Si}_{\text{rad}}$	1σ	$\delta^{30}\text{Si}_{\text{spo}}$	1σ	Place of analysis	$\delta^{30}\text{Si}_{\text{sponges}}$	2σ	
58-2-142	502.87	350.77			0.16	0.03	IFREMER Brest	0.02	0.91	15
58-3-20	503.15	346.32	0.47	0.06	0.18	0.03	IFREMER Brest	-0.18	0.92	11
58-3-50	503.45	342.02	0.45	0.05	0.29	0.09	IFREMER Brest	-0.15	0.81	6
58-3-140	504.35	324.22	0.71	0.10	-0.06	0.07	IFREMER Brest	-0.38	0.68	8
58-4-90	505.35	305.77						-0.56	0.93	9
58-4-101	505.46	304.93	0.37	0.06	0.25	0.23	IFREMER Brest	-0.22	0.86	9
58-4-120	505.65	299.04	0.46	0.04	-0.05	0.11	IFREMER Brest	-0.24	0.61	12
59-1-10	506.20	289.21	0.45	0.05	0.29	0.09	IFREMER Brest	-0.62	0.71	12
59-1-70	506.80	277.52	-0.46	0.05	0.44	0.03	IFREMER Brest	-0.10	0.85	11
59-1-110	507.20	269.73	0.41	0.01	-0.12	0.05	IFREMER Brest	-0.16	0.77	9
					-0.18	0.09	IFREMER Brest			
					-0.15	0.10				
59-1-121	507.31	267.59	0.27	0.02	-0.58	0.18	IFREMER Brest	0.12	0.44	10
59-2-70	508.30	248.30	0.28	0.01	0.44	0.12	IFREMER Brest	0.20	0.67	9
59-2-130	508.90	236.61	0.25	0.04	-0.18	0.09	IFREMER Brest	-0.36	0.82	11
59-3-10	509.20	230.77	0.16	0.03	-0.12	0.07	IFREMER Brest	0.43	0.90	13
59-3-50	509.60	223.26	0.32	0.05			IFREMER Brest	-0.08	0.65	9
59-3-90	510.00	199.26	0.50	0.06			IFREMER Brest	-0.10	0.62	11
59-3-126	510.36	177.39	0.40	0.00			IFREMER Brest	0.99	1.03	8
59-CC-29		177.39	0.42	0.13			IFREMER Brest	1.18	0.31	6
60-1-30	511.14	94.18	0.62	0.03	-0.11	0.13	IFREMER Brest	0.26	0.91	10
60-1-40	511.24	91.07	0.84	0.03	0.45	0.03	IFREMER Brest			
60-1-50	511.34	87.44	0.65	0.04	0.26	0.11	IFREMER Brest	0.33	0.70	10
60-1-60	511.44	83.41	0.75	0.04	0.49	0.03	IFREMER Brest			
60-1-70	511.54	76.48	0.45	0.06			IFREMER Brest	0.16	0.88	10

60-1-80	511.64	69.98	0.96	0.04	0.07	0.05	IFREMER Brest			
60-1-90	511.74	64.94	0.90	0.22			IFREMER Brest	0.12	0.69	15
			0.85	0.02			IFREMER Brest			
			<u>0.88</u>	<u>0.22</u>						
60-1-100	511.84	60.97	0.51	0.01	-0.05	0.03	IFREMER Brest			
60-1-105	511.89	59.30	0.77	0.01	0.29	0.03	IFREMER Brest			
60-1-110	511.94	57.78						0.98	0.73	9
60-1-115	511.99	56.25	0.78	0.02	0.16	0.01	IFREMER Brest			
60-1-120	512.04	53.98	0.57	0.05			IFREMER Brest			
60-1-125	511.90	51.07	0.49	0.01	0.62	0.03	IFREMER Brest			
60-1-130	512.14	48.15	0.51	0.04			IFREMER Brest	1.19	0.71	10
			0.70	0.02			IFREMER Brest			
			<u>0.60</u>	<u>0.05</u>						
60-1-135	512.19	45.34	0.62	0.06			IFREMER Brest			
60-1-140	512.24	40.78	0.67	0.03			IFREMER Brest			
60-1-145	512.29	36.82	0.78	0.02			IFREMER Brest			
60-1-149	512.33	31.69						0.12	0.80	12
60-2-8	512.41	26.54			0.32	0.00	IFREMER Brest			
60-2-20	512.54	18.28	0.21	0.07	0.54	0.07	IFREMER Brest	0.51	0.51	10
			0.56	0.01			IFREMER Brest			
			<u>0.38</u>	<u>0.07</u>			IFREMER Brest			
60-2-23	512.57	16.42	0.52	0.05			IFREMER Brest			
60-2-35	512.69	8.54	0.69	0.10			Vegacenter Stockholm			
60-2-40	512.74	6.19	0.67	0.14	0.43	0.19	Vegacenter Stockholm	0.31	0.91	10
			0.55	0.00			IFREMER Brest			
			<u>0.61</u>	<u>0.14</u>						
60-2-45	512.79	4.46	0.83	0.10			Vegacenter Stockholm			
60-2-50	512.84	2.61	0.99	0.09	0.54	0.11	Vegacenter Stockholm			
60-2-53	512.87	1.45	1.05	0.07			Vegacenter Stockholm			
60-2-56	512.90	0.35	1.02	0.18	0.44	0.12	Vegacenter Stockholm			
60-2-58	512.92	-0.57	1.18	0.06			Vegacenter Stockholm			
60-2-60	512.94	-1.72	0.94	0.03	0.36	0.05	IFREMER Brest	0.12	0.59	12
60-2-62	512.96	-2.87			0.26	0.12	Vegacenter Stockholm			
60-2-65	512.99	-4.72	1.00	0.07			Vegacenter Stockholm			

60-2-69	513.03	-7.24	0.75	0.08	0.41	0.09	Vegacenter Stockholm			
60-2-76	513.10	-11.34	1.24	0.07			Vegacenter Stockholm			
60-2-80	513.14	-13.46	1.03	0.06	0.20	0.18	Vegacenter Stockholm			
60-2-92	513.26	-20.01	1.22	0.22			Vegacenter Stockholm			
60-2-95	513.29	-21.85	1.23	0.13	0.34	0.00	Vegacenter Stockholm			
60-2-99	513.33	-24.31	1.04	0.10	0.28	0.11	Vegacenter Stockholm			
60-2-104	513.38	-27.38	0.95	0.03			Vegacenter Stockholm			
60-2-110	513.44	-31.11						0.29	0.65	11
60-2-114	513.48	-33.59	1.06	0.12			Vegacenter Stockholm			
60-2-120	513.54	-37.54	1.12	0.11			Vegacenter Stockholm			
60-2-125	513.59	-40.86	0.82	0.07			Vegacenter Stockholm			
60-2-130	513.64	-44.20	1.11	0.10	0.15	0.22	Vegacenter Stockholm			
60-2-135	513.69	-47.52	1.16	0.00	0.53	0.18	Vegacenter Stockholm			
60-2-140	513.74	-50.13	1.41	0.17			Vegacenter Stockholm			
60-2-145	513.79	-54.47	1.25	0.06	0.33	0.19	Vegacenter Stockholm			
60-3-1	513.85	-59.41	1.18	0.26	0.25	0.14	Vegacenter Stockholm			
60-3-5	513.89	-64.13	0.95	0.17			Vegacenter Stockholm			
60-3-10	513.94	-70.28	0.59	0.15	0.34	0.14	IFREMER Brest	0.77	0.75	9
			0.63	0.04			IFREMER Brest			
			0.61	0.15						
60-3-13	515.14	-131.12			-0.20	0.07	IFREMER Brest			
60-3-70	515.71	-164.22						-0.29	0.87	12
60-3-130	516.31	-199.06						0.50	0.64	10
60-4-29	516.63	-217.64	0.71	0.12			IFREMER Brest	0.93	0.51	8
60-4-132	517.66	-277.45						0.34	0.73	10
60-5-30	518.35	-317.51	0.75	0.08			IFREMER Brest	1.69	0.87	8
61-1-70	518.75	-340.74	0.98	0.11			IFREMER Brest	1.06	0.72	10
61-3-90	521.95	-526.55	1.49	0.07			IFREMER Brest	1.07	0.81	10
Secondary reference material		n	$\delta^{30}\text{Si}$	1σ	$\delta^{29}\text{Si}$	1σ				
Big Batch		2	-10.37	0.04	-5.31	0.04				
Diatomite		2	1.3	0.08	0.66	0.04				