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Palacín Lizarbe, Carlos; Camarero, Lluís; Catalan, Jordi. Denitrification temperature dependence in remote, cold, and N-poor lake sediments. DOI 10.1002/2017WR021680

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Denitrification temperature dependence in remote, cold and N-poor lake sediments

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Table S1. Experimental results. Denitrifying activity rate (r_{dT}) ($\text{N}_2\text{O } \mu\text{mol m}^{-2} \text{h}^{-1}$) for each experimental temperature (T) ($^{\circ}\text{C}$), and Q_{10} and apparent activation energy (E_a) (kJ mol^{-1}) for each nitrate enrichment level. Sampling date (YYMMDD). E_a^* calculated with a temperature range of 5-10 $^{\circ}\text{C}$ instead of 5-15 $^{\circ}\text{C}$.

Date and lake	Core	Sensor	Initial NO_3^- (μM)	$7\mu\text{M NO}_3^-$ added				$14\mu\text{M NO}_3^-$ added				$28\mu\text{M NO}_3^-$ added					
				r_{d5}	r_{d15}	Q_{10}	E_a	r_{d5}	r_{d15}	Q_{10}	E_a	r_{d5}	r_{d15}	Q_{10}	E_a	r_{d10}	E_a^*
130903 Redon	A	1	5									7.1	17.4	2.4	59	14.4	92
	B	2	6									12.2	24.0	2.0	45	21.5	74
	C	3	6									8.5	19.8	2.3	57	15.4	79
	D	4	6									13.0	27.9	2.2	51	22.6	72
	E	5	6									9.0	17.0	1.9	42	15.1	68
131007 Redon	F	1	6					2.9	10.5	3.6	86						
	G	2	6					2.7	10.0	3.7	87						
	H	3	5					4.9	16.9	3.4	82						
	I	4	5					3.8	10.7	2.9	70						
	J	5	6					5.7	15.0	2.6	65						
131104 Redon	K	1	5					3.4	15.2	4.5	100	8.5	30.2	3.5	84		
	L	2	5	1.7	9.3	5.3	111	8.3	16.2	1.9	44	15.5	35.6	2.3	56		
	M	3	6	2.6	7.5	2.9	70	5.9	13.4	2.3	55	11.3	29.7	2.6	64		
	N	4	4	2.9	8.6	2.9	72	10.6	17.0	1.6	31	12.8	49.7	3.9	91		
	O	5	5					6.9	15.0	2.2	51	18.2	60.5	3.3	80		
131111 Plan	P	1	1					3.2	11.5	3.6	86	5.7	21.3	3.7	88		
	Q	2	2	3.6	10.0	2.8	69	4.6	17.3	3.8	89	17.9	30.1	1.7	34		
	R	3	1	0.5	4.6	9.3	149	2.6	8.8	3.4	81	7.5	23.8	3.2	76		
	S	4	1	4.3	7.3	1.7	35	3.8	20.3	5.3	112	25.2	37.1	1.5	26		
	T	5	1	1.0	2.2	2.2	51	2.4	9.5	4.0	92	9.6	21.1	2.2	52		
131118 Llong	U	1	7	2.1	10.7	5.2	110	2.2	12.2	5.6	115	6.2	13.9	2.3	54		
	V	2	8	1.3	6.1	4.8	104	3.9	7.7	2.0	45	8.5	14.1	1.7	34		
	W	3	9	1.1	2.6	2.3	57	2.2	3.5	1.6	31						
	X	4	9	2.7	6.4	2.4	57	8.2	15.5	1.9	42	12.9	29.3	2.3	55		
	Y	5	8	2.9	4.0	1.4	22	3.9	7.7	2.0	45	4.2	9.9	2.3	57		

Table S2. Studies on the temperature dependence of the denitrification rates in aquatic ecosystems

Study reference	T range	Ecosystem	Habitat	Experimental device	Method	NO ₃ ⁻ <i>in situ</i> (μM)	NO ₃ ⁻ added (μM)	Q ₁₀	E _a (kJ mol ⁻¹)	E _a (eV)	E _a change (%)	Experiment al T (°C)	<i>In situ</i> T (°C)
This study (experimental)	Complete	Lake (N-poor)	Sediment	Sediment core	Acetylene inhibition	5.1 (0.8-9.3)	7	3.6 (1.4-9.3)	76 (22-149)	0.79 (0.23-1.54)		10 (5-15)	5
This study (experimental)	Complete	Lake (N-poor)	Sediment	Sediment core	Acetylene inhibition	5.1 (0.8-9.3)	14	3.1 (1.6-5.6)	70 (31-114)	0.73 (0.32-1.18)		10 (5-15)	5
This study (experimental)	Complete	Lake (N-poor)	Sediment	Sediment core	Acetylene inhibition	5.1 (0.8-9.3)	28	2.5 (1.5-3.9)	58 (26-91)	0.6 (0.27-0.94)		10 (5-15)	5
This study (experimental) ^a	Complete	Lake (N-poor)	Sediment	Sediment core	Acetylene inhibition	5.3 (5.1-5.9)	28	2.2 (1.9-2.4)	51 (42-59)	0.53 (0.44-0.61)	51	10 (5-15)	5
This study (experimental) ^a	<i>In situ</i>	Lake (N-poor)	Sediment	Sediment core	Acetylene inhibition	5.3 (5.1-5.9)	28	3.3 (2.8-4.1)	77 (68-92)	0.8 (0.71-0.95)	51	7.5 (5-10)	5
Messer and Brezonik [1984]	Complete	Lake	Sediment	Slurries	Acetylene inhibition	-	12500	2.6	70	0.73		25 (14-36)	25 ^b
Myrstener et al. [2016]	Complete	Lake (N-poor)	Sediment	Slurries	Acetylene inhibition	5	62.7	1.77	47	0.49	13	14.5 (4-25)	4
Myrstener et al. [2016]	<i>In situ</i>	Lake (N-poor)	Sediment	Slurries	Acetylene inhibition	5	62.7	NA	53	0.55	13	7 (4-10)	4
Cavari and Phelps [1977]	Complete	Lake	<i>P. aeruginosa</i> culture	Culture	Nitrate removal	(7-107)	143	1.4	26	0.27	23	22.5 (15-30)	16 (16-30) ^c
Cavari and Phelps [1977]	<i>In situ</i>	Lake	<i>P. aeruginosa</i> culture	Culture	Nitrate removal	(7-107)	143	1.6	32	0.33	23	18.5 (15-22)	16 (16-30) ^c
Veraart et al. [2011]	Complete	Pond	Sediment	Microcosms	¹⁵ N-Tracer	-	119	8.6	179	1.86		18 (11-25)	17.5
Veraart et al. [2011]	Complete	Pond	Sediment	Ditch enclosure	¹⁵ N-Tracer	-	50	11.8	155	1.61		14 (8-20)	14 ^d
Boulêtreau et al. [2012]	Complete	Stream	River biofilm	Slurries	Acetylene inhibition	85.7	1714	7 (4.2-9.8)	137 (91-183)	1.42 (0.94-1.9)	-69	16 (1-31)	7.2
Boulêtreau et al. [2012]	<i>In situ</i>	Stream	River biofilm	Slurries	Acetylene inhibition	85.7	1714	NA	43	0.45	-69	6.5 (1-12)	7.2
Holmes et al. [1996]	Complete	Stream	Sediment (Parafluvial)	Slurries	Acetylene inhibition	5.1	14286	3.5	88	0.91		17 (10-24)	23 (17-29)
Pfenning and McMahon [1997]	Complete	River (eutrophic)	Sediment	Slurries	Acetylene inhibition	-	357	1.8	41	0.43		13 (4-22)	10 (2-18.1)
Silvennoinen et al. [2008]	Complete	River (eutrophic)	Sediment	Sediment core	¹⁵ N-Tracer	-	30	3.1	82	0.85		12.5 (5-20)	17
Pattinson et al. [1998]	Complete	River (eutrophic)	Sediment	Sediment core	Acetylene inhibition	2300	814	2.2	48	0.5	167	12.5 (5-20)	5
Pattinson et al. [1998]	<i>In situ</i>	River (eutrophic)	Sediment	Sediment core	Acetylene inhibition	2300	814	7.1	128	1.33	167	7.5 (5-10)	5

Study reference	T range	Ecosystem	Microbial assemblage	Sample	Method	NO ₃ ⁻ <i>in situ</i> (μM)	NO ₃ ⁻ added (μM)	Q ₁₀	E _a (kJ mol ⁻¹)	E _a (eV)	E _a change (%)	Estimation (°C)	<i>In situ</i> T (°C)
<i>Pattinson et al. [1998]</i>	Complete	River (eutrophic)	Sediment	Sediment core	Acetylene inhibition	2300	1528	2	45	0.47	109	12.5 (5-20)	5
<i>Pattinson et al. [1998]</i>	<i>In situ</i>	River (eutrophic)	Sediment	Sediment core	Acetylene inhibition	2300	1528	4.2	94	0.97	109	7.5 (5-10)	5
<i>Pattinson et al. [1998]</i>	Complete	River (eutrophic)	Sediment	Sediment core	Acetylene inhibition	2300	2242	1.7	33	0.34	115	12.5 (5-20)	5
<i>Pattinson et al. [1998]</i>	<i>In situ</i>	River (eutrophic)	Sediment	Sediment core	Acetylene inhibition	2300	2242	2.9	71	0.74	115	7.5 (5-10)	5
<i>Pattinson et al. [1998]</i>	Complete	River (eutrophic)	Sediment	Sediment core	Acetylene inhibition	2300	2957	1.6	29	0.3	110	12.5 (5-20)	5
<i>Pattinson et al. [1998]</i>	<i>In situ</i>	River (eutrophic)	Sediment	Sediment core	Acetylene inhibition	2300	2957	2.5	61	0.63	110	7.5 (5-10)	5
<i>Sheibley et al. [2003]</i>	Complete	Hyporheic zone	Sediment	Sediment core	Nitrate removal	0.14	no add	5.3	101	1.05		15 (8-22)	15 ^d
<i>Jørgensen et al. [2009]</i>	Complete	Groundwater aquifer	Sediment	Slurries	Nitrate removal	-	521	1.9	42	0.44	69	19 (9-29)	9
<i>Jørgensen et al. [2009]</i>	<i>In situ</i>	Groundwater aquifer	Sediment	Slurries	Nitrate removal	-	521	2.7	71	0.74	69	11.5 (9-14)	9
<i>Ambus [1993]</i>	Complete	Riparian (stream-land)	Soil (riparian, stream side)	Slurries	Acetylene inhibition	-	14286	2.9	65	0.67	18	12.5 (2-23)	7.5 (0-15)
<i>Ambus [1993]</i>	<i>In situ</i>	Riparian (stream-land)	Soil (riparian, stream side)	Slurries	Acetylene inhibition	-	14286	NA	77	0.8	18	8.5 (2-15)	7.5 (0-15)
<i>Westermann and Ahring [1987]</i>	Complete	Swamp	Sediment Alder swamp	Slurries	Acetylene inhibition	12.5	1000	2.9 (2.8-3.0)	73 (70-76)	0.76 (0.73-0.79)	32	13.5 (2-25)	7.5 (0-15)
<i>Westermann and Ahring [1987]</i>	<i>In situ</i>	Swamp	Sediment Alder swamp	Slurries	Acetylene inhibition	12.5	1000	NA	96	1	32	6 (2-10)	7.5 (0-15)
<i>King and Nedwell [1984]</i>	Complete	Wetland (Salt-marsh)	Culture dominated by <i>Pseudomonas</i> spp.	Culture	Nitrate removal	-	1000	NA	98	1.02		8 (3-13)	15 (0-20) ^e
<i>King and Nedwell [1984]</i>	Complete	Wetland (Salt-marsh)	Culture dominated by <i>Vibrio</i> spp.	Culture	Nitrate removal	-	1000	NA	60	0.62		18.5 (6-31)	15 (0-20) ^f
<i>Brin et al. [2017]</i>	Complete	Estuary (June)	Sediment	Slurries	Acetylene inhibition	1.1 (0.6-2.1) ^h	100 ⁱ	NA	36 ^k	0.37	25	17 (3-31)	16
<i>Brin et al. [2017]</i>	<i>In situ</i>	Estuary (June)	Sediment	Slurries	Acetylene inhibition	1.1 (0.6-2.1) ^h	100 ⁱ	NA	45	0.46	25	16 (12-20)	16
<i>Brin et al. [2017]</i>	Complete	Estuary (August)	Sediment	Slurries	Acetylene inhibition	1.1 (0.6-2.1) ^h	100 ⁱ	NA	46 ^k	0.48	78	15 (3-27)	22
<i>Brin et al. [2017]</i>	<i>In situ</i>	Estuary (August)	Sediment	Slurries	Acetylene inhibition	1.1 (0.6-2.1) ^h	100 ⁱ	NA	82	0.85	78	21.5 (18-25)	22
<i>Brin et al. [2017]</i>	Complete	Estuary (January)	Sediment	Slurries	Acetylene	1.1	100 ⁱ	NA	53 ^k	0.55	56	15	6

Study reference	T range	Ecosystem	Microbial assemblage	Sample	Method	NO ₃ ⁻ <i>in situ</i> (μM)	NO ₃ ⁻ added (μM)	Q ₁₀	E _a (kJ mol ⁻¹)	E _a (eV)	E _a change (%)	Estimation T (°C)	<i>In situ</i> T (°C)
					inhibition	(0.6-2.1) ^h						(3-27)	
<i>Brin et al. [2017]</i>	<i>In situ</i>	Estuary (January)	Sediment	Slurries	Acetylene inhibition	1.1 (0.6-2.1) ^h	100 ⁱ	NA	83	0.86	56	6.5 (3-10)	6
<i>Brin et al. [2017]</i>	Complete	Marine, temperate (January)	Sediment (continental shelf)	Slurries	Acetylene inhibition	1.5 (1.0-4.0) ^h	100 ⁱ	NA	60 ^k	0.63	9	19 (3-35)	6
<i>Brin et al. [2017]</i>	<i>In situ</i>	Marine, temperate (January)	Sediment (continental shelf)	Slurries	Acetylene inhibition	1.5 (1.0-4.0) ^h	100 ⁱ	NA	66	0.69	9	6.5 (3-10)	6
<i>Brin et al. [2017]</i>	Complete	Marine, temperate (June)	Sediment (continental shelf)	Slurries	Acetylene inhibition	1.5 (1.0-4.0) ^h	100 ⁱ	NA	44 ^k	0.45	70	15 (3-27)	11
<i>Brin et al. [2017]</i>	<i>In situ</i>	Marine, temperate (June)	Sediment (continental shelf)	Slurries	Acetylene inhibition	1.5 (1.0-4.0) ^h	100 ⁱ	NA	74	0.77	70	11 (8-14)	11
<i>Brin et al. [2017]</i>	Complete	Marine, temperate (July)	Sediment (continental shelf)	Slurries	Acetylene inhibition	1.5 (1.0-4.0) ^h	100 ⁱ	NA	51 ^k	0.52	-65	15 (3-27)	16
<i>Brin et al. [2017]</i>	<i>In situ</i>	Marine, temperate (July)	Sediment (continental shelf)	Slurries	Acetylene inhibition	1.5 (1.0-4.0) ^h	100 ⁱ	NA	18	0.18	-65	16 (12-20)	16
<i>Brin et al. [2017]</i>	Complete	Marine, temperate (September)	Sediment (continental shelf)	Slurries	Acetylene inhibition	1.5 (1.0-4.0) ^h	100 ⁱ	NA	39 ^k	0.4	-17	13.5 (3-24)	17
<i>Brin et al. [2017]</i>	<i>In situ</i>	Marine, temperate (September)	Sediment (continental shelf)	Slurries	Acetylene inhibition	1.5 (1.0-4.0) ^h	100 ⁱ	NA	32	0.33	-17	17.5 (14-21)	17
<i>Brin et al. [2017]</i>	Complete	Marine, temperate (March)	Sediment (continental shelf)	Slurries	Acetylene inhibition	1.5 (1.0-4.0) ^h	100 ⁱ	NA	44 ^k	0.45	6	13.5 (3-24)	7
<i>Brin et al. [2017]</i>	<i>In situ</i>	Marine, temperate (March)	Sediment (continental shelf)	Slurries	Acetylene inhibition	1.5 (1.0-4.0) ^h	100 ⁱ	NA	46	0.48	6	6.5 (3-10)	7
<i>Brin et al. [2017]</i>	Complete	Marine, temperate (March)	Sediment (microcosm at 4°C, 2 weeks)	Slurries	Acetylene inhibition	-	100 ⁱ	NA	41 ^k	0.43		12.5 (3-22)	4 ^j
<i>Brin et al. [2017]</i>	Complete	Marine, temperate (March)	Sediment (microcosm at 4°C, 12 weeks)	Slurries	Acetylene inhibition	-	100 ⁱ	NA	40 ^k	0.42		12.5 (3-22)	4 ^j
<i>Brin et al. [2017]</i>	Complete	Marine, temperate (March)	Sediment (microcosm at 4°C + C, 12 weeks)	Slurries	Acetylene inhibition	-	100 ⁱ	NA	37 ^k	0.38		12.5 (3-22)	4 ^j
<i>Brin et al. [2017]</i>	Complete	Marine, temperate (March)	Sediment (microcosm at 17°C, 12 weeks)	Slurries	Acetylene inhibition	-	100 ⁱ	NA	44 ^k	0.46		12.5 (3-22)	17 ^j
<i>Brin et al. [2017]</i>	Complete	Marine, temperate (March)	Sediment (microcosm at 17°C + C, 12 weeks)	Slurries	Acetylene inhibition	-	100 ⁱ	NA	46 ^k	0.48		12.5 (3-22)	17 ^j
<i>Canion et al. [2014]</i>	Complete	Marine, temperate (March)	Sediment (Near shore, permeable)	Slurries	¹⁵ N-Tracer	53.9	100	2.5	74	0.77	41	13 (0-26)	5.6
<i>Canion et al. [2014]</i>	<i>In situ</i>	Marine, temperate (March)	Sediment (Near shore, permeable)	Slurries	¹⁵ N-Tracer	53.9	100	NA	104	1.08	41	5 (0-10)	5.6
<i>Canion et al. [2014]</i>	Complete	Marine, temperate (June)	Sediment (Near shore, permeable)	Slurries	¹⁵ N-Tracer	0.8	100	2	54	0.56	-26	13 (0-26)	17.9

<i>Canion et al. [2014]</i>	<i>In situ</i>	Marine, temperate (June)	Sediment (Near shore, permeable)	Slurries	¹⁵ N-Tracer	0.8	100	NA	40	0.41	-26	18 (17-19)	17.9
Study reference	T range	Ecosystem	Microbial assemblage	Sample	Method	NO₃⁻ <i>in situ</i> (μM)	NO₃⁻ added (μM)	Q₁₀	E_a (kJ mol⁻¹)	E_a (eV)	E_a change (%)	Estimationl T (°C)	<i>In situ</i> T (°C)
<i>Kraft et al. [2014]</i>	Complete	Marine, temperate	Bact. com. isolation (sandy tidal flat)	Culture	¹⁵ N-Tracer	-	1000	1.4	22	0.23	18	20 (10-30)	11 (2-19)
<i>Kraft et al. [2014]</i>	<i>In situ</i>	Marine, temperate	Bact. com. isolation (sandy tidal flat)	Culture	¹⁵ N-Tracer	-	1000	NA	26	0.27	18	12.5 (10-15)	11 (2-19)
<i>Rysgaard et al. [2004]</i>	Complete	Marine, polar	Sediment (polar 40 m water depth)	Slurries	¹⁵ N-Tracer	3.8	50	NA	61	0.63	110	11.5 (-2-25)	0 (-0.5-4)
<i>Rysgaard et al. [2004]</i>	<i>In situ</i>	Marine, polar	Sediment (polar 40 m water depth)	Slurries	¹⁵ N-Tracer	3.8	50	NA	128	1.33	110	-0.8 (-2-0.5)	0 (-0.5-4)
<i>Canion et al. [2014]</i>	Complete	Marine, polar	Sediment (Near shore, permeable)	Slurries	¹⁵ N-Tracer	0.5	100	2.3	61	0.63	34	10 (-1-21)	6.8
<i>Canion et al. [2014]</i>	<i>In situ</i>	Marine, polar	Sediment (Near shore, permeable)	Slurries	¹⁵ N-Tracer	0.5	100	NA	82	0.85	34	7.5 (6-9)	6.8
<i>Canion et al. [2014]</i>	Complete	Marine, subtropical gulf	Sediment (Near shore, permeable)	Slurries	¹⁵ N-Tracer	1.7	100	3.8	100	1.04	30	17.5 (-1-36)	29.9
<i>Canion et al. [2014]</i>	<i>In situ</i>	Marine, subtropical gulf	Sediment (Near shore, permeable)	Slurries	¹⁵ N-Tracer	1.7	100	NA	130	1.35	30	29.5 (27-32)	29.9
<i>Canion et al. [2014]</i>	Complete	Marine, subtropical bay	Sediment (Near shore, permeable)	Slurries	¹⁵ N-Tracer	2.2	100	5	121	1.25	47	19 (3-35)	31.8
<i>Canion et al. [2014]</i>	<i>In situ</i>	Marine, subtropical bay	Sediment (Near shore, permeable)	Slurries	¹⁵ N-Tracer	2.2	100	NA	191	1.98	47	32 (30-34)	31.8
<i>Cameron and Schipper [2010]</i>	Complete	Artificial	DEN bed (sawdust (<i>Pinus radiata</i>))	DEN bed	Nitrate removal	-	10714	2.4	63	0.65		19 (14-24)	19 ^d
<i>Warneke et al. [2011]</i>	Complete	Artificial	DEN bed (wood chips and sawdust of <i>P. radiata</i>)	DEN bed	Nitrate removal	-	8929	2.1	53	0.54		19.5 (15-24)	19.5 ^d
<i>Cameron and Schipper [2010]</i>	Complete	Artificial	DEN bed (woodchips of <i>Pinus radiata</i>)	DEN bed	Nitrate removal	-	10714	1.9	45	0.47		19 (14-24)	19 ^d
<i>Cameron and Schipper [2010]</i>	Complete	Artificial	DEN bed (woodchips of <i>Eucalyptus</i>)	DEN bed	Nitrate removal	-	10714	2	50	0.52		19 (14-24)	19 ^d
<i>Cameron and Schipper [2010]</i>	Complete	Artificial	DEN bed (maize cobs)	DEN bed	Nitrate removal	-	10714	1.3	16	0.17		19 (14-24)	19 ^d
<i>Cameron and Schipper [2010]</i>	Complete	Artificial	DEN bed (wheat straw)	DEN bed	Nitrate removal	-	10714	1.2	14	0.15		19 (14-24)	19 ^d
<i>Cameron and Schipper [2010]</i>	Complete	Artificial	DEN bed (green waste)	DEN bed	Nitrate removal	-	10714	1.2	11	0.12		19 (14-24)	19 ^d

Notes (Table S2):

Numerical values in parenthesis indicate the range, minimum and maximum values.

Temperature (T) range: “complete” refers to the full experimental temperature range in the paper; “*in situ*” refers to a shorter range closer to the *in situ* temperature.

Q₁₀ calculation: $Q_{10} = \frac{r_{dj}}{r_{di}} \left(\frac{10}{T_j - T_i} \right)$, where r_d is the denitrification rate and T the absolute temperature.

E_a (kJ mol⁻¹) calculation from: $\frac{r_{dj}}{r_{di}} = \exp\left[\frac{E_a}{R} \left(\frac{1}{T_i} - \frac{1}{T_j}\right)\right]$. Where R is the gas constant (8.314 J K⁻¹ mol⁻¹), T the absolute temperature and r_d the denitrification rate.

E_a (eV) from E_a (kJ mol⁻¹) data: E_a (eV) = 0.01037 * E_a (kJ mol⁻¹).

E_a change (%) = $\left(\frac{E_a \text{ in situ } T \text{ range} - E_a \text{ complete } T \text{ range}}{E_a \text{ complete } T \text{ range}} \right) * 100$.

^a Lake Redon, field campaign 3rd September 2013 (see Table S1).

^b Annual mean temperature (air) (<http://www.worldclim.org>).

^c Lake Kinneret water temperature: hypolimnion 16°C (constant), epilimnion 16-30°C [Gal *et al.*, 2003].

^d Experimental mean temperature.

^e Culture isolated at 10°C.

^f Culture isolated at 25°C.

^h Nitrate *in situ* is mean porewater nitrate (μM) data from Brin *et al.* [2014].

ⁱ Nitrate added: 100 nmol NO₃⁻N mL sediment⁻¹.

^j Sediments collected in the field at 7°C.

^k Mean E_a values of 45, 48 and 42 kJ mol⁻¹ for estuary, continental shelf and microcosms samples, respectively [Brin *et al.*, 2017], used in Fig. 3 plot.

Regression models relating the DEN E_a (kJ mol^{-1}) to the inverse of the nitrate concentration ($[\text{NO}_3^-]^{-1}$) (μM):

In this section, we provide a brief introduction of the linear mixed-effects models and detailed information about the performed alternative regression models relating the DEN E_a (kJ mol^{-1}) to the inverse of the nitrate concentration ($[\text{NO}_3^-]^{-1}$) (μM). The aim is to clarify and complement the information in the main manuscript (section 3.1 and Table 2).

Many common statistical models can be expressed as linear models that incorporate both fixed effects, which are parameters associated with an entire population or with certain repeatable levels of experimental factors, and random effects, which are associated with individual experimental units drawn at random from a population. A model with both fixed effects and random effects is called a mixed-effects model [Pinheiro and Bates, 1978]. All the performed regression models in this study have the same fixed part with the inverse of the nitrate concentration ($[\text{NO}_3^-]^{-1}$) (μM) in the overlying water of the lake sediments (Table 2).

Mixed-effects models are primarily used to describe relationships between a response variable and some covariates in data that are grouped according to one or more classification factors. Examples of such grouped data include longitudinal data, repeated measures data, multilevel data, and block designs. By associating common random effects to observations sharing the same level of a classification factor, mixed-effects models flexibly represent the covariance structure induced by the grouping of the data [Pinheiro and Bates, 1978].

The model 0 and 1 are simple linear regression models, without any random effects, they differ in the function used to fit the model. In model 0 is used the *lm* function from the R package *stats*, the design was inspired by the *S* function of the same name described in Chambers [1992]. Model 1 uses the *gls* function, from the R package *nlme* [Pinheiro et al., 2007], which fits linear models using generalized least squares. Both models are identical, have the same coefficient and intercept. We develop the model 1 to compare it with model 2, which incorporate the temporal autocorrelation intrinsic to sequential experimental additions, i.e., if it is the first, second or third addition in the core sample. Model 2 uses the same *gls* function to fit the model but incorporates an auto-regressive model of order 1 (corAR1) accounting for auto-correlation (see further details in Zuur et al. [2009] section 6.1). We conclude that temporal-autocorrelation was not interfering with the results as model 2 does not improve model 1. The two models explain the same fixed and global variance, and an ANOVA comparing the models showed that the more complex model 2 did not improve significantly ($p=0.56$) model 1. Models 3-10 are all mixed-effects models with the same fixed part with the inverse of the nitrate concentration and different composition of the random part. Models 3, 4 and 5 account for the sensor particular performance (#1, #2, #3, #4 or #5, see Table S1). Model 4 just modify the slope, not the intercept. Models 5 (nested in the sensor), 6 and 7 consider the core (sample) effect. Model 8 takes into account the three nitrate enrichment levels, and model 9 the addition order (first, second and third). Finally, model 10 takes into account the lake effect. Models with more than 1 factor crossed in the random part (not shown) were built with the *lmer* function within the *lme4* R package [Bates et al., 2015], these models did not improve model 1, 3 or 4 ($p(>0.05)$ in ANOVAs and have higher AICc values). As we mention in the manuscript (Results 3.1), the models accounting for the sensor effects (model 3 and 4 in Table 2) showed the lower AICc values, explained more variance and were the only ones that improved the initial model 1, which did not have the random part (ANOVA p-values of 0.047 and

0.008 for model 3 and 4, respectively). We selected model 4 as the best estimation because it is simpler than 3.

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