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Lanthanum anomalies as fingerprints of methanotrophy

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Supplementary Information

The Supplementary Information includes:

- Methods
- Tables S-1 to S-9

Methods

Once on board the ship, the bivalves were sampled by dissecting pieces of soft tissues and shell from the same organism. The soft tissues were rinsed with deionised water to remove residual seawater, and freeze dried. Shells were scrapped to remove traces of soft tissues and sediments, rinsed with deionised water and dried. For each sample, about 100 mg were spiked with a solution of pure Tm and digested in a Teflon beaker by HNO_3 (carbonate), sequentially by HNO_3/H_2O_2 and HNO_3 (soft tissue), or by HF/HNO_3, HNO_3 and HCl (sediments). REEs have been separated from the major elements and concentrated before analysis (Barrat *et al.*, 1996, 2020). Abundances in most samples were determined using a high-resolution inductively coupled plasma-mass spectrometer Thermo Element XR at Institut Universitaire Européen de la Mer (IUEM), Plouzané, France. Each sample was analysed in duplicate or in triplicate, and the results were averaged (Barrat *et al.*, 2016). Results on a carbonate standard obtained during the sessions are given in Table S-9. Clam shells from Nankai Trough were analysed using a similar procedure at Laboratoire de Géodynamique des Chaines Alpines (Université Joseph Fourier, Grenoble) using a Fisons PQ2+ instrument.

The La and Ce anomalies are calculated using the La/La* and Ce/Ce* ratios, where La* and Ce* are the extrapolated La and Ce concentration for a smooth Post Archean Australian Shale-normalised REE pattern and X_{sn} is the concentration of element X normalised to PAAS: $La_{sn}^* = Pr_{sn}^3/Nd_{sn}^2$ and $Ce_{sn}^* = Pr_{sn}^2/Nd_{sn}$. Based on standards and sample replicates, the precisions for abundances and element ratios are usually much better than 5 % (2 RSD).

Table S-1 Samples and location of the sampling localities.

Site	latitude longitude	Cruise	Sampling date	Depth	Sample ID	Species	Chemotrophic symbionts
Nankai Trough	33.66°N 137.90°E	Kaiko-Nankai Nautile manned submersible	08-08-1989 to 09-10-1989	3848 m	KN1-(2), KN1-(A)	*Calyptogena sp.*	Thiotrophs
				2170 m	KN2	*Calyptogena sp.*	Thiotrophs
				3787 m	KN7	*Calyptogena sp.*	Thiotrophs
				2200 m	KN14-(2)	*Calyptogena sp.*	Thiotrophs
Site F	22.12°N 119.29°E	*Jiaolong* manned submersible *ROPOS* ROV	6-18-2013 5-13-2018	1120 m	BA1 to 3	*Bathylomodius aduloides*	Thiotrophs
				1120 m	GP1 to 10	*Gigantidas platifrons*	Methanotrophs
Haima	16.73°N 110.48°E	*Haima* ROV	5-30-2018	1390 m	GH1 to 5	*Gigantidas haimaensis*	Methanotrophs
			5-30-2018	1390 m	CM1 to 2	*Calyptogena marissinica*	Thiotrophs
Ouessant	48.4522°N 5.0971°W		feb-18	0	Ou1 to 4	*Mytilus edulis*	none
Chausey	48.8886°N 1.7863°W		nov-19	0	Ch1 to 3	*Mytilus edulis*	none
Gouville/Mer	49.0916°N 1.6197°W		nov-19	0	Go1 to 3	*Mytilus edulis*	none

Table S-2 REE and Y abundances (in ng/g) in *Gigantidas platifrons* from site F (S : shell ; F: foot; G : gill ; H: haslet; M : mantle; n.a.: not analysed).

	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Ce/Ce*	La/La*
Shells																
GP1, S	25.8	68.3	14.4	2.68	9.00	1.67	0.46	2.19	0.32	1.87	0.43	1.17	0.94	0.16	0.56	4.83
GP2, S	22.1	55.7	11.2	2.72	9.95	1.81	0.45	2.14	0.28	1.69	0.38	1.06	0.91	0.15	0.47	4.62
GP3, S	26.5	109	24.6	3.88	12.71	2.11	0.53	2.37	0.35	2.03	0.45	1.24	1.03	0.18	0.65	5.09
GP4, S	19.6	38.3	14.0	2.21	7.55	1.52	0.40	2.00	0.28	1.73	0.39	1.14	1.06	0.17	0.68	3.42
GP5, S	27.2	49.9	13.8	1.92	6.37	1.24	0.35	1.82	0.30	2.06	0.53	1.59	1.55	0.27	0.74	4.77
GP6, S	20.8	35.7	16.2	2.01	6.29	1.13	0.31	1.63	0.25	1.63	0.39	1.16	1.10	0.19	0.78	2.92
GP7, S	44.8	62.3	23.3	3.69	12.68	2.60	0.73	3.87	0.57	3.54	0.82	2.36	2.06	0.34	0.68	3.35
GP8, S	14.5	33.4	10.1	1.59	5.36	0.91	0.24	1.23	0.18	1.11	0.26	0.76	0.66	0.11	0.67	4.00
GP9, S	14.1	45.9	12.7	1.78	5.28	0.73	0.19	1.05	0.15	0.93	0.22	0.63	0.54	0.09	0.66	3.80
GP10, S	32.3	68.8	19.2	3.33	11.08	1.99	0.53	2.79	0.42	2.72	0.65	1.97	1.91	0.32	0.60	3.85
Gills																
GP1, G	19.2	2322	185	26.2	36.3	3.22	0.71	n.a.	0.44	2.47	0.52	1.33	1.10	0.19	0.31	2.87
GP2, G	34.0	1817	208	37.0	58.5	4.61	0.96	n.a.	0.58	3.49	0.79	2.28	2.12	0.37	0.28	2.07
GP3, G	32.8	1709	161	28.1	52.2	5.42	1.13	n.a.	0.63	3.59	0.81	2.37	2.20	0.38	0.33	3.53
GP4, G	25.4	1974	177	36.2	52.2	3.16	0.63	n.a.	0.38	2.29	0.54	1.63	1.58	0.28	0.22	1.91
GP5, G	28.2	2027	226	31.5	50.6	4.33	0.90	n.a.	0.53	2.99	0.66	1.86	1.67	0.29	0.36	2.80
GP6, G	36.1	2225	384	41.2	59.9	5.21	1.18	n.a.	0.79	4.49	0.97	2.58	2.03	0.34	0.42	1.92
GP7, G	26.4	2025	311	44.4	67.1	5.04	1.06	n.a.	0.68	3.81	0.81	2.18	1.79	0.30	0.33	1.75
GP8, G	21.7	1680	218	30.4	33.7	3.02	0.95	n.a.	0.53	2.95	0.63	1.64	1.32	0.22	0.25	1.14
GP9, G	34.0	2312	329	62.1	122	10.2	2.15	n.a.	1.10	5.49	1.06	2.55	1.85	0.30	0.33	2.44
GP10, G	22.8	917	158	20.9	41.8	5.69	1.10	n.a.	0.64	3.31	0.65	1.72	1.59	0.27	0.47	2.94

Table S-2 (continued) REE and Y abundances (in ng/g) in *Gigantidas platifrons* from site F (S : shell ; F: foot; G : gill ; H: haslet; M : mantle).

	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Ce/Ce*	La/La*
Mantles																
GP1, M	24.4	49.7	8.02	1.60	5.35	1.05	0.28	1.61	0.24	1.72	0.45	1.42	1.34	0.22	0.53	5.86
GP2, M	33.7	143	26.4	4.12	11.4	2.07	0.56	3.03	0.43	2.82	0.69	2.04	1.84	0.30	0.56	4.51
GP3, M	27.5	81.2	19.7	3.60	13.1	2.44	0.59	2.82	0.38	2.36	0.56	1.65	1.37	0.22	0.62	5.01
GP4, M	34.1	137	26.1	4.59	13.0	2.19	0.55	2.88	0.42	2.72	0.66	2.05	1.89	0.31	0.50	4.07
GP5, M	29.1	43.1	12.8	2.06	7.17	1.42	0.38	2.10	0.32	2.18	0.56	1.75	1.51	0.24	0.67	4.27
GP6, M	38.7	94.3	30.8	4.52	13.8	2.31	0.59	3.17	0.45	2.95	0.73	2.23	1.97	0.32	0.65	3.26
GP7, M	18.4	150	40.6	5.50	15.4	3.17	0.61	2.89	0.38	2.24	0.50	1.33	0.89	0.14	0.65	3.62
GP8, M	7.0	42.8	13.3	2.05	7.20	1.22	0.27	1.19	0.15	0.82	0.18	0.51	0.42	0.069	0.71	4.36
GP9, M	20.8	117	39.4	6.14	21.0	3.55	0.85	4.00	0.47	2.32	0.42	0.96	0.69	0.10	0.69	3.74
GP10, 10	12.6	70.7	19.6	2.80	8.36	1.41	0.34	1.62	0.23	1.39	0.31	0.92	0.85	0.14	0.65	3.78
Feet																
GP5, F	9.20	41.2	11.5	1.46	4.30	0.83	0.21	1.16	0.15	0.96	0.22	0.64	0.56	0.10	0.73	4.14
GP7, F	4.76	47.7	12.4	1.61	4.17	0.70	0.14	0.71	0.085	0.52	0.12	0.35	0.33	0.065	0.63	3.39
Haslets																
GP2, H	21.6	255	64.7	9.7	27.1	4.72	1.07	4.83	0.63	3.44	0.66	1.77	1.87	0.31	0.59	3.52
GP7, H	113	547	217	29.0	95.3	20.6	4.95	20.6	3.12	17.7	3.45	9.48	9.45	1.34	0.77	3.42

Table S-3 REE and Y abundances (in ng/g) in *Bathylomodius aduloides* from Site F (S: shell; G: gill; M: mantle).

	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Ce/Ce*	La/La*
Shells																
BA1, S	61.8	48.7	44.7	7.67	33.0	6.74	1.74	8.49	1.07	5.97	1.22	3.21	2.61	0.41	0.78	1.98
BA2, S	60.9	64.0	51.7	8.24	33.7	5.94	1.49	7.66	0.92	4.80	0.96	2.26	1.38	0.21	0.80	2.19
BA3, S	56.1	57.1	43.4	7.34	30.1	5.29	1.30	6.67	0.84	4.60	0.94	2.31	1.63	0.26	0.76	2.20
Gill																
BA2, G	242	71.1	85.6	16.3	90.2	34.6	7.82	36.6	4.86	26.5	5.27	13.8	13.5	2.42	0.91	2.26
Mantle																
BA2, M	151	88.6	139	22.4	117	34.5	9.29	42.4	5.48	28.3	5.20	11.6	6.56	0.91	1.02	1.83

Table S-4 REE and Y abundances (in ng/g) in *Gigantidas Haimaensis* from Haima (S: shell; F: foot; G: gill; M: mantle).

	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Ce/Ce*	La/La*
Shells																
GH1, S	25.9	77.4	51.5	4.97	15.3	2.32	0.52	3.03	0.35	2.10	0.47	1.31	1.20	0.19	1.00	2.50
GH2, S	25.2	41.2	26.8	3.12	10.8	2.11	0.50	2.76	0.35	2.08	0.46	1.30	1.19	0.19	0.93	2.65
GH3, S	43.3	51.9	24.7	2.77	9.75	2.20	0.62	3.65	0.52	3.42	0.83	2.52	2.52	0.43	0.98	3.92
GH4, S	28.1	27.9	16.4	1.97	6.77	1.41	0.37	2.25	0.31	2.05	0.49	1.47	1.38	0.23	0.90	2.84
GH5, S	27.1	61.3	29.0	3.36	11.1	1.93	0.48	2.82	0.34	2.12	0.49	1.39	1.25	0.20	0.89	3.35
Mantles																
GH1, M	63.0	281	247	25.2	83.9	15.1	3.02	15.3	1.73	9.40	1.85	4.81	3.73	0.52	1.02	2.08
GH2, M	9.40	63.8	27.6	2.83	7.19	1.93	0.30	1.62	0.18	0.94	0.17	0.41	0.37	0.059	0.78	2.45
GH3, M	57.9	265	182	14.0	36.6	6.54	1.53	8.85	1.03	6.07	1.31	3.48	2.48	0.33	1.07	2.19
GH4, M	27.4	115	73.3	6.80	20.3	4.01	0.84	4.27	0.53	2.92	0.58	1.47	1.19	0.18	1.01	2.53
GH5, M	33.0	141	95.7	7.84	21.3	3.60	0.89	4.83	0.65	4.11	0.98	2.86	2.27	0.34	1.03	2.22
Feet																
GH1, F	31.8	219	161	15.7	48.2	7.88	1.58	8.62	0.89	4.84	0.97	2.54	2.05	0.30	0.98	2.21
GH2, F	5.32	39.4	17.7	1.57	4.06	0.81	0.15	0.86	0.10	0.61	0.13	0.36	0.39	0.068	0.91	2.81
GH3, F	15.8	114	73.4	5.36	12.8	1.86	0.42	2.86	0.27	1.71	0.37	1.04	0.95	0.15	1.02	2.04
GH4, F	10.2	51.7	33.1	2.78	7.39	1.16	0.27	1.49	0.18	1.12	0.25	0.66	0.61	0.098	0.99	2.22
GH5, F	11.2	67.4	42.4	3.26	7.84	1.16	0.25	1.41	0.18	1.23	0.29	0.89	0.93	0.16	0.98	2.01
Gills																
GH2, G	23.6	1827	446	53.7	77.0	6.24	1.17	7.39	0.57	3.03	0.59	1.52	1.39	0.25	0.37	1.18
GH3, G	47.5	840	374	34.4	69.4	9.76	1.98	14.2	1.15	5.91	1.09	2.63	2.30	0.36	0.69	1.67
GH4, G	39.3	862	263	31.4	58.3	9.13	1.59	8.68	0.91	4.51	0.80	1.85	1.60	0.26	0.49	1.60
GH5, G	37.9	773	285	23.4	44.2	6.12	1.25	7.18	0.79	4.48	0.95	2.55	2.36	0.40	0.72	1.99

Table S-5 REE and Y abundances (in ng/g) in *Calyptogena marissinica* from Haima (S: shell; G: gill; M: mantle).

	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Ce/Ce*	La/La*
Shells																
CM1, S	3.55	5.43	10.1	1.23	4.67	0.95	0.19	0.89	0.10	0.59	0.11	0.28	0.23	0.031	0.975	1.073
CM2, S	20.5	27.9	57.4	6.62	25.8	5.28	1.10	5.07	0.63	3.37	0.64	1.65	1.38	0.19	1.06	1.08
Mantles																
CM1, M	849	743	1087	154	627	139	32.2	154	20.5	111	21.5	54.2	40.5	5.80	0.90	1.34
CM2, M	1352	1215	1534	231	941	203	46.6	220	27.8	128	20.1	43.3	29.6	3.91	0.84	1.47
Gills																
CM1, G	211	154	86.7	22.1	93.5	20.2	5.18	27.6	3.41	18.5	3.75	9.23	6.25	0.93	0.52	2.10
CM2, G	888	706	463	102	422	85.3	20.9	111	13.9	76.5	15.8	39.5	27.9	4.44	0.59	2.00

Table S-6 REE abundances in *Calyptogena* shells (in ng/g) from Nankai Trough (n.a: not analysed).

	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Ce/Ce*	La/La*
KN1-A	n.a.	58.7	68.7	8.84	30.02	5.45	1.35	6.61	0.99	5.36	1.01	2.47	1.56	0.21	0.83	1.29
KN1-(2)	n.a.	1474	2487	224	814	128	30.66	139	19.13	102	18.26	40.76	22.94	2.97	1.26	1.46
KN2	n.a.	51.9	33.0	5.29	19.93	3.65	0.94	4.30	0.54	2.70	0.55	1.35	0.76	0.12	0.73	2.34
KN7	n.a.	250	442	41.9	140	22.60	5.13	25.10	3.60	19.65	3.80	9.04	5.78	0.78	1.10	1.13
KN14-(2)	n.a.	51.5	75.5	8.11	27.87	5.27	1.02	5.90	0.97	5.75	1.22	3.28	2.49	0.33	1.00	1.27

Table S-7 REE and Y concentrations (in ng/g) in *Mytilus edulis* (S: shell; ST: soft tissue) from Brittany (Ouessant) and Normandy (Chausey and Gouville/Mer), France.

	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Ce/Ce*	La/La*
Ouessant																
Ou1, S	7.54	5.69	4.89	0.80	3.62	0.84	0.20	1.19	0.13	0.66	0.12	0.25	0.10	0.012	0.86	2.43
Ou2, S	5.97	7.72	6.00	1.03	3.93	0.79	0.17	1.06	0.10	0.50	0.092	0.20	0.11	0.015	0.70	1.84
Ou3, S	6.35	6.76	5.81	0.90	3.56	0.73	0.17	1.02	0.11	0.54	0.096	0.21	0.087	0.014	0.80	2.00
Ou4, S	7.11	6.93	5.62	0.88	3.69	0.76	0.17	1.07	0.11	0.57	0.11	0.25	0.13	0.017	0.84	2.33
Ou1, ST	23.7	51.6	59.0	6.79	25.6	4.67	0.99	4.68	0.57	3.13	0.61	1.59	1.38	0.23	1.02	1.82
Ou2, ST	18.3	46.6	46.6	5.34	20.3	4.10	0.82	3.91	0.49	2.54	0.47	1.17	0.98	0.15	1.04	2.13
Ou4, ST	39.2	66.0	64.4	8.14	33.9	7.10	1.57	7.84	0.98	5.20	1.02	2.59	2.15	0.34	1.03	2.38
Chausey																
Ch1, S	12.6	9.89	16.1	1.99	8.28	1.69	0.39	2.06	0.23	1.16	0.20	0.46	0.33	0.044	1.05	1.45
Ch2, S	10.4	8.47	11.9	1.56	6.53	1.28	0.29	1.57	0.18	0.97	0.19	0.48	0.34	0.048	1.00	1.60
Ch3, S	10.7	8.48	11.4	1.49	6.34	1.20	0.29	1.55	0.18	0.99	0.20	0.49	0.30	0.040	1.02	1.74
Gouville/Mer																
Go1, S	9.12	8.31	12.3	1.60	6.55	1.36	0.31	1.57	0.19	1.03	0.20	0.48	0.33	0.044	0.99	1.48
Go2, S	13.3	9.48	13.3	1.78	7.57	1.59	0.39	2.08	0.24	1.23	0.23	0.52	0.32	0.044	0.99	1.62
Go3, S	8.93	8.82	10.9	1.37	5.74	1.12	0.26	1.44	0.17	0.90	0.18	0.42	0.25	0.035	1.03	1.89

Table S-8 REE and Y abundances (µg/g) in terrigeneous sediments from Haima, Site F, and results for USGS basalt BCR2.

	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Ce/Ce*	La/La*
Site F, sediments																
LN1	20.93	33.67	69.66	7.75	28.70	5.35	1.09	4.59	0.64	3.63	0.72	1.99	1.91	0.276	1.04	1.00
LN10	22.10	36.89	74.77	8.33	31.26	5.86	1.17	4.95	0.69	3.86	0.76	2.12	2.01	0.291	1.05	1.05
Haima, sediments																
mussel zone	23.21	32.52	67.09	7.53	28.16	5.40	1.09	4.79	0.69	3.95	0.78	2.21	2.10	0.302	1.04	1.02
clam zone	21.24	28.29	56.99	6.47	24.10	4.61	0.94	4.21	0.60	3.43	0.70	1.95	1.82	0.263	1.03	1.02
BCR2 (three distinct dissolutions)																
#1	38.08	25.80	54.53	6.91	29.07	6.60	1.93	6.83	1.06	6.43	1.32	3.65	3.36	0.490	1.04	1.11
#2	37.99	25.86	54.54	6.96	29.19	6.65	1.94	6.87	1.06	6.42	1.33	3.66	3.38	0.492	1.03	1.10
#3	37.92	24.84	52.70	6.82	28.75	6.58	1.90	6.72	1.05	6.39	1.31	3.64	3.40	0.486	1.02	1.09
average (n=3)	**38.00**	**25.50**	**53.92**	**6.90**	**29.00**	**6.61**	**1.92**	**6.81**	**1.06**	**6.41**	**1.32**	**3.65**	**3.38**	**0.489**	**1.03**	**1.10**
RSD (%)	*0.21*	*2.25*	*1.97*	*0.99*	*0.78*	*0.57*	*1.05*	*1.14*	*0.64*	*0.30*	*0.48*	*0.32*	*0.45*	*0.66*	*1.00*	*1.10*
Jochum *et al.* (2016)	36.07	25.08	53.12	6.83	28.26	6.55	1.99	6.81	1.08	6.42	1.31	3.67	3.39	0.50	1.01	1.06

Table S-9 REE and Y abundances REE+Y abundances (in ng/g) in Cal-S standard obtained during the course of this study.

	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Ce/Ce*	La/La*
average, n=41	**2177**	**806**	**313**	**89.2**	**363**	**63.7**	**15.9**	**92.3**	**13.9**	**99.6**	**26.3**	**82.0**	**67.1**	**10.3**	**0.45**	**2.52**
RSD (%)	*3.58*	*1.33*	*2.72*	*1.70*	*1.70*	*1.68*	*1.33*	*1.41*	*1.23*	*1.29*	*1.12*	*1.11*	*1.23*	*1.22*	*1.23*	*0.80*
Le Goff *et al.* (2019)	2065	793	302	87.1	359	62.4	15.5	91.6	13.7	98.3	26.0	81.0	66.3	10.16	0.45	2.62
Potts *et al.* (2000)	1944	787	333	90	357	64	16	93	14	100	26	81	68	11	0.46	2.63

Supplementary Information References

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