

1
2 **Supplementary Information**

3
4 **for:**

5
6 **Compound-specific recording of gadolinium**
7 **pollution in coastal waters by great scallops**
8

9 **Samuel Le Goff¹, Jean-Alix Barrat^{1*}, Laurent Chauvaud², Yves-Marie**
10 **Paulet², Bleuenn Gueguen³, Douraied Ben Salem⁴**

11
12 ¹Laboratoire Géosciences Océan (UMR CNRS 6538), Université de Bretagne Occidentale et
13 Institut Universitaire Européen de la Mer, Place Nicolas Copernic, 29280 Plouzané, France.

14 ²Laboratoire des Sciences de l'Environnement Marin (UMR CNRS 6539), LIA BeBEST,
15 Université de Bretagne Occidentale et Institut Universitaire Européen de la Mer, Place
16 Nicolas Copernic, 29280 Plouzané, France.

17 ³UMS CNRS 3113, Université de Bretagne Occidentale et Institut Universitaire Européen de la
18 Mer, Place Nicolas Copernic, 29280 Plouzané, France.

19
20 ⁴LaTIM (UMR INSERM 1101) Université de Bretagne Occidentale. 22, avenue C. Desmoulins,
21 29238 Brest Cedex 3, France

22

23 **1/ Sampling areas**

24

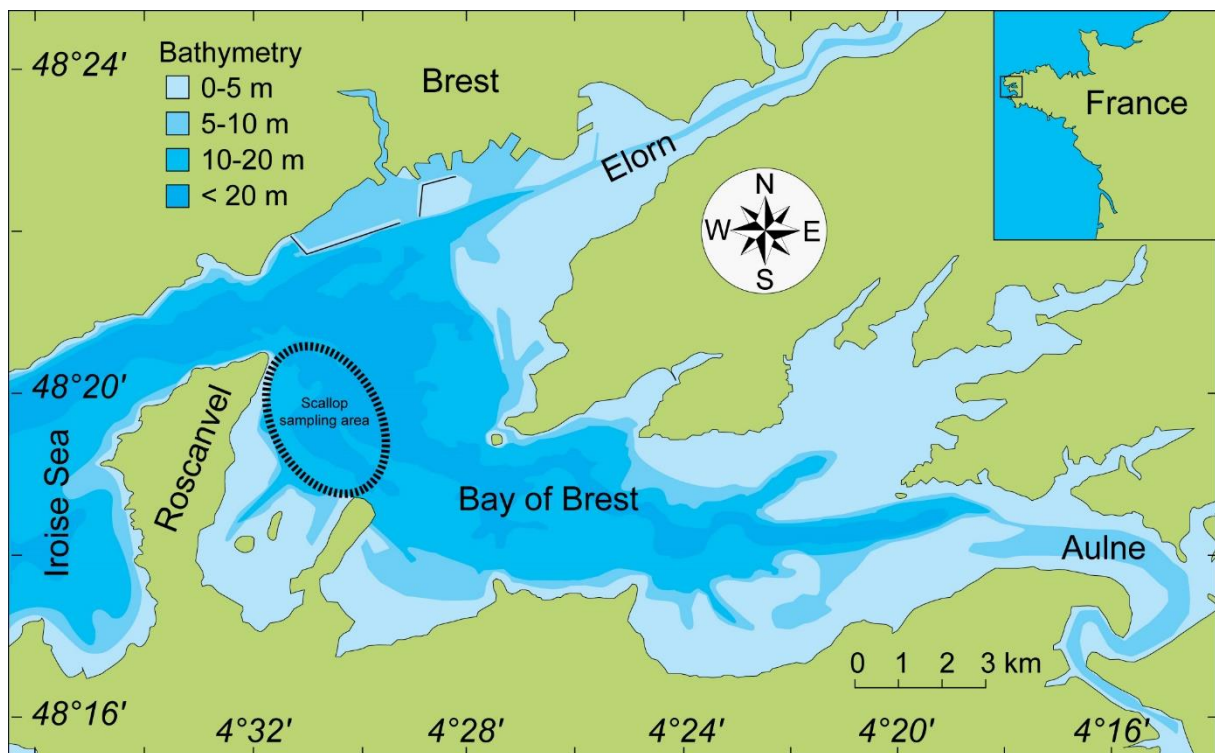
25 **a/ Great scallops**

26

27 Bay of Brest is a semi-enclosed marine ecosystem connected to shelf waters by a 2 km-
28 narrow and 40 m-deep strait (Supplementary figure 1). Two rivers, the Elorn and the Aulne,
29 make up 80% of the total freshwater input in the Bay. All the scallops were fished near
30 Roscanvel at depth > 20 m. In this area, the contribution of freshwater is limited. Bottom water
31 salinity is quite stable from spring to fall (34-35 ‰) and decreases only to 32.5 ‰ during flood
32 tide in winter (S1).

33

34



35

36

37 Figure S1. Location of the sampling sites in Bay of Brest (circled area).

38

39

40 **b/ Limpets**

41

42 We analysed limpets from Bay of Brest and from Fuerteventura (Canary Islands). In both cases,
43 shellfish were collected alive.

44

45 Bay of Brest: Limpets PMOUL3 and 4 were sampled in February 2018 in the “Moulin Blanc”
46 beach, near Brest. Limpet PM3 was collected the same month on the shore of the island of the
47 dead (“Ile des Morts”), located south of the scallop sampling area.

48

49 Fuerteventura: samples were collected in November 2017 in the south of the island.

50

51 **2/ Sample preparation**

52

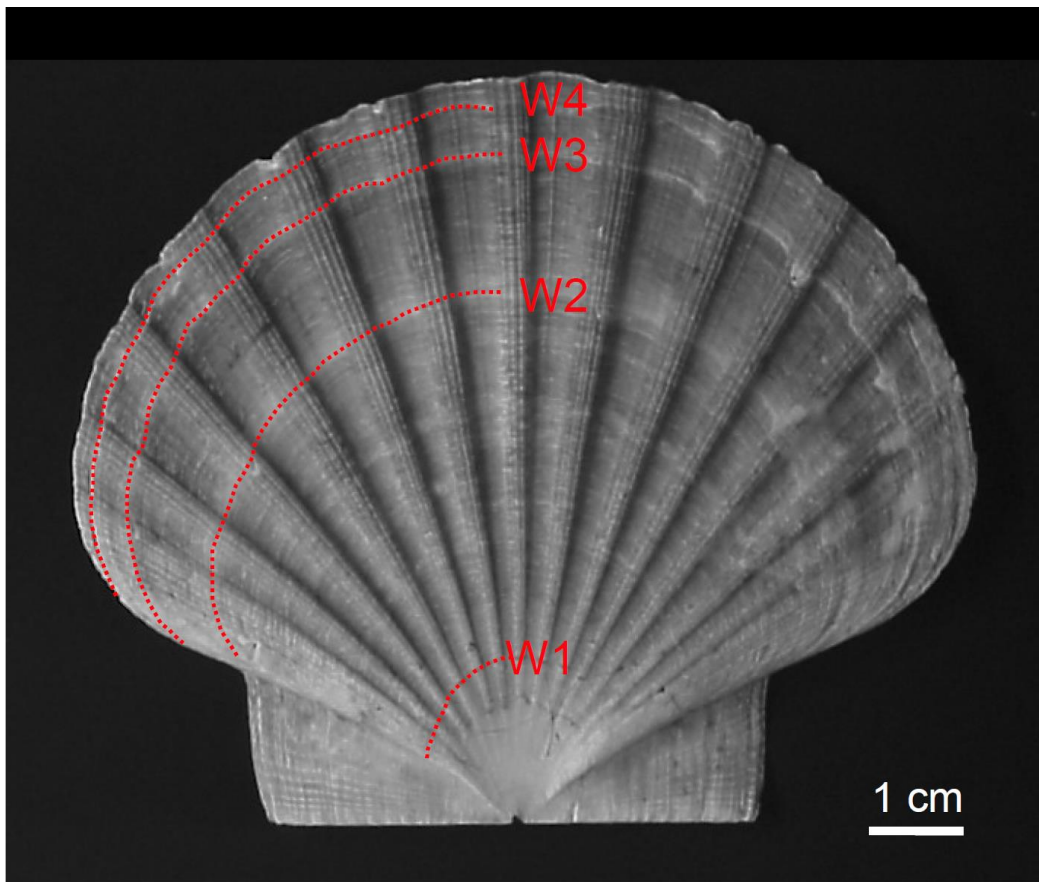
53 The left valves of the scallops were abraded with an electric milling cutter to remove any
54 adhering material from the surface, and fragments of their last growth rind were removed (Fig.

55 S2). They were then rinsed with ultrapure water and dried.

56

57

58



59

60

61

62 Figure S2. Upper surface of the left valve of a great scallop (adult). “Winter marks” (W1 to
63 W4) deposited during spring growth restart are clearly visible, allowing an unambiguous aging.

64

65

66

67

68

69

70

71

72 **3/ Results for the Cal-S standard and shells**

73

74

75

76

77 Three tables follow:

78

79

80 Supplementary table 1- results for a carbonate standard.

81

82 Supplementary table 2- results for scallop shells.

83

84 Supplementary table 3- results for limpet shells.

85 Supplementary table 1. REE+Y abundances (in ng/g) in the Meuse limestone (prepared by the Service d'Analyses de Roches et des Minéraux
 86 (SARM), Nancy) obtained during the course of this study. Each analysis (#1 to 11) was obtained on a separated dissolution.
 87
 88

	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Gd/Gd*	ΔGd
#1	1992	764	295	84.4	349	61.0	15.02	88.7	13.34	95.9	25.28	79.0	64.4	9.93	1.17	12.8
#2	2005	773	301	85.4	353	61.8	15.16	90.0	13.56	97.4	25.67	80.0	64.6	9.89	1.17	12.9
#3	2004	771	291	84.5	349	61.0	15.04	88.9	13.40	96.3	25.51	79.1	64.8	10.02	1.17	12.8
#4	2099	815	314	89.6	369	64.1	15.89	92.9	13.89	99.9	26.36	81.8	65.7	9.90	1.17	13.6
#5	2087	813	304	88.5	365	63.1	15.70	92.9	13.80	99.0	26.17	81.3	66.0	10.00	1.18	14.3
#6	2060	800	294	86.9	360	62.3	15.51	91.3	13.66	98.2	26.02	81.0	66.3	10.08	1.18	13.6
#7	2065	790	314	91.1	359	63.0	15.64	91.8	13.86	99.0	26.34	82.3	67.6	10.48	1.17	13.1
#8	2120	814	306	88.9	367	63.7	15.84	94.0	13.98	99.6	26.38	82.1	67.4	10.30	1.18	14.6
#9	2111	799	308	87.2	361	62.7	15.57	92.7	13.87	99.5	26.38	82.0	67.9	10.45	1.18	14.0
#10	2068	786	293	85.4	353	61.6	15.44	91.5	13.70	98.2	25.93	80.8	67.0	10.36	1.18	14.0
#11	2101	794	300	86.6	359	62.5	15.61	92.3	13.82	98.7	26.17	81.5	67.2	10.35	1.18	13.9
average	2065	793	302	87.1	359	62.4	15.49	91.6	13.72	98.3	26.02	81.0	66.3	10.16	1.17	13.6
RSD (%)	2.2	2.2	2.6	2.5	2.0	1.7	1.9	1.9	1.5	1.4	1.5	1.4	1.9	2.3	0.5	4.6
Potts (S2)	1944	787	333	90	357	64	16	93	14	100	26	81	68	11	1.17	13.3

90 Supplementary table 2. REE+Y abundances (ng/g) in scallop shells from Bay of Brest. All the analyses were obtained on the last growth rind of
 91 different shells.

#	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Gd/Gd*	ΔGd
1960-A	25.25	51.71	91.31	11.30	40.68	6.35	1.28	4.89	0.63	3.32	0.60	1.48	0.96	0.13	1.05	0.24
1989-A	27.12	19.26	25.12	3.20	12.53	2.45	0.63	3.04	0.43	2.46	0.51	1.30	0.59	0.10	1.16	0.42
1989-B	61.50	65.33	108	13.80	54.09	10.47	2.30	10.64	1.43	7.83	1.54	3.99	2.87	0.44	1.12	1.11
1990-A	30.56	22.93	35.43	4.35	17.35	3.64	0.87	4.63	0.59	3.22	0.63	1.62	0.85	0.13	1.24	0.90
1990-B	28.79	23.38	35.48	4.48	16.93	3.39	0.78	4.24	0.53	2.92	0.59	1.53	0.83	0.14	1.26	0.88
1991-A	32.99	27.20	39.55	4.95	18.77	3.21	0.74	3.82	0.54	3.15	0.65	1.71	1.02	0.14	1.13	0.45
1991-B	33.11	24.38	36.19	4.53	17.94	3.48	0.79	3.97	0.56	3.22	0.66	1.68	1.04	0.15	1.12	0.43
1992-A	53.26	58.94	105	13.86	52.82	10.11	2.03	8.39	1.07	5.80	1.15	2.99	1.83	0.25	1.08	0.61
1993-A	36.32	34.05	57.08	7.21	28.78	6.26	1.19	5.87	0.77	4.28	0.84	2.20	1.45	0.21	1.11	0.58
1993-B	29.82	25.61	38.25	4.96	19.58	3.97	0.84	4.82	0.58	3.33	0.66	1.66	1.11	0.16	1.27	1.03
1994-B	32.11	24.04	57.85	4.60	19.57	3.56	0.71	4.54	0.55	3.23	0.64	1.66	1.00	0.14	1.28	1.00
1995-A	34.84	57.48	97.45	12.29	46.96	8.33	1.36	6.98	0.78	4.07	0.74	1.84	1.07	0.14	1.18	1.08
1995-B	45.75	40.90	66.72	8.47	34.07	7.18	1.47	8.15	1.05	5.65	1.08	2.73	1.77	0.26	1.19	1.33
1996-A	25.70	17.42	25.28	3.23	12.72	2.66	0.55	3.73	0.48	2.60	0.52	1.37	0.84	0.12	1.28	0.81
1996-B	46.27	67.01	120	14.13	47.76	8.38	1.52	8.04	1.07	6.00	1.18	3.16	2.20	0.31	1.10	0.76
1997-A	38.35	30.48	42.86	5.36	21.28	3.86	0.86	5.20	0.67	3.87	0.79	1.98	1.17	0.17	1.27	1.09
1997-B	29.16	26.14	36.75	4.58	18.95	3.60	0.83	4.65	0.56	3.21	0.62	1.61	1.03	0.15	1.30	1.09
1998-A	46.24	34.98	52.55	6.47	25.46	4.99	1.18	6.37	0.81	4.65	0.94	2.41	1.27	0.20	1.25	1.27
1998-B	55.84	45.10	72.88	8.87	34.82	7.30	1.72	9.65	1.17	6.55	1.28	3.31	2.13	0.32	1.31	2.26
1999-A	50.48	41.65	63.66	7.76	29.72	5.74	1.27	6.82	0.92	5.21	1.07	2.72	1.66	0.24	1.17	1.01
1999-B	54.47	50.34	84.12	10.76	43.20	8.67	1.87	9.60	1.23	6.85	1.33	3.53	2.46	0.36	1.19	1.53
2000-A	31.79	27.49	46.50	5.92	22.95	4.75	1.09	5.82	0.72	3.87	0.75	1.97	1.30	0.19	1.26	1.20
2000-B	32.51	25.29	42.65	5.62	22.10	4.62	1.04	5.63	0.70	3.85	0.77	2.01	1.33	0.21	1.25	1.14

92

93

94 Supplementary table 2 (continue).

#	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Gd/Gd*	Δ Gd
2001-A	44.87	40.04	59.48	7.18	28.12	5.65	1.30	7.00	0.87	4.82	0.95	2.46	1.47	0.22	1.26	1.43
2001-B	38.56	34.15	47.18	5.74	21.44	4.20	0.93	5.48	0.65	3.62	0.72	1.88	0.96	0.15	1.32	1.31
2002-A	81.18	89.86	157	20.50	81.70	16.56	3.56	16.73	2.18	11.85	2.28	5.97	4.56	0.69	1.14	2.01
2002-B	54.23	53.55	91.81	12.15	48.43	9.87	2.11	10.74	1.34	7.42	1.41	3.66	2.65	0.40	1.20	1.81
2003-A	45.83	41.48	63.60	7.96	31.46	6.17	1.22	7.21	0.89	5.00	0.98	2.46	1.52	0.21	1.24	1.40
2003-B	47.23	39.04	61.87	8.09	31.58	6.53	1.35	7.61	0.97	5.57	1.10	2.81	1.83	0.26	1.21	1.31
2004-A	35.26	23.51	30.81	3.93	15.20	2.81	0.65	4.64	0.51	2.86	0.59	1.47	0.56	0.09	1.50	1.54
2005-B	53.10	76.21	153	18.08	66.39	12.17	2.47	10.27	1.49	8.19	1.53	3.94	3.05	0.41	1.00	-0.01
2006-A	105	108	189	24.92	99.58	20.37	4.05	19.29	2.75	15.46	2.99	7.90	6.59	0.98	1.05	0.89
2006-B	41.18	36.07	54.04	7.08	27.92	5.54	1.17	6.84	0.81	4.50	0.89	2.32	1.64	0.24	1.29	1.56
2007-A	33.46	41.95	79.59	9.59	35.52	6.64	1.57	5.80	0.79	4.45	0.86	2.30	1.83	0.28	1.05	0.29
2007-B	48.87	58.46	111	13.69	52.54	10.38	2.46	9.73	1.33	7.21	1.36	3.53	2.71	0.39	1.08	0.69
2008-A	45.06	36.15	54.45	6.99	28.25	5.78	1.25	7.11	0.92	5.13	1.01	2.56	1.60	0.28	1.22	1.30
2008-B	33.61	23.47	34.63	4.31	17.36	4.25	0.80	4.96	0.60	3.53	0.70	1.83	1.24	0.20	1.25	0.99
2009-A	36.58	43.72	84.40	10.32	38.47	7.21	1.75	6.20	0.85	4.85	0.93	2.51	2.05	0.30	1.04	0.23
2009-B	33.20	52.30	105	13.12	48.99	8.87	2.03	6.75	0.88	4.76	0.89	2.33	1.91	0.28	1.03	0.22
2010-A	72.66	80.81	150	18.27	68.54	13.05	3.11	11.75	1.71	9.67	1.90	5.06	4.01	0.57	1.02	0.22
2010-B	87.58	102	185	22.94	84.40	15.79	3.67	14.21	1.99	11.10	2.17	5.75	4.46	0.63	1.04	0.61
2011-A	51.55	50.99	87.97	10.54	39.12	7.24	1.74	7.09	0.99	5.67	1.14	3.07	2.31	0.33	1.08	0.52
2011-B	77.07	81.23	150	17.89	66.61	12.23	2.94	10.93	1.59	9.10	1.82	4.94	3.82	0.55	1.02	0.17
2012-A	33.49	22.47	30.53	4.01	16.37	3.18	0.68	4.87	0.54	3.36	0.64	1.69	0.97	0.15	1.45	1.52
2012-B	60.57	65.57	119	14.47	53.38	9.82	2.28	8.82	1.25	7.06	1.40	3.72	2.90	0.42	1.03	0.29
2013-A	68.65	62.78	97.25	12.18	46.81	9.04	2.15	9.61	1.32	7.38	1.45	3.73	2.58	0.36	1.12	1.01

95

96

97 Supplementary table 2 (continue)

#	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Gd/Gd*	Δ Gd
2013-B	76.14	83.78	149	18.14	67.84	12.68	3.06	11.64	1.62	9.21	1.80	4.76	3.70	0.53	1.05	0.59
2014-A	60.84	64.91	113	13.81	55.67	10.23	2.32	9.98	1.35	7.70	1.43	3.73	2.71	0.38	1.10	0.90
2014-B	37.03	41.95	76.07	9.35	34.80	6.52	1.57	6.16	0.82	4.66	0.90	2.41	1.92	0.31	1.09	0.53
2015-A	20.75	21.17	40.36	5.04	18.95	3.69	0.88	3.67	0.49	2.75	0.52	1.41	1.15	0.17	1.12	0.39
2015-B	34.16	39.57	76.69	9.41	35.03	6.62	1.65	5.98	0.81	4.51	0.86	2.31	1.92	0.28	1.07	0.39
2017-A	27.96	29.26	53.77	6.65	24.73	4.67	1.15	4.49	0.59	3.41	0.65	1.72	1.37	0.20	1.11	0.44
2017-B	51.62	30.26	43.87	5.57	22.06	4.85	1.20	7.08	1.05	6.48	1.31	3.39	2.34	0.33	1.18	1.08
2018-JM1	21.38	14.51	15.92	2.13	8.87	1.89	0.44	2.51	0.34	1.92	0.36	0.95	0.80	0.10	1.21	0.44
2018-JM3	43.86	29.08	36.63	4.54	18.26	3.71	0.81	5.33	0.70	3.94	0.74	2.17	1.71	0.20	1.28	1.16
2018-SA1	11.31	6.80	7.06	1.01	4.20	0.92	0.22	1.47	0.18	0.93	0.18	0.42	0.20	-	1.39	0.41
2018-A	40.98	36.47	45.80	5.20	19.61	3.55	0.91	4.60	0.63	3.38	0.69	1.71	0.90	0.13	1.20	0.76
2018-B	40.21	33.86	50.25	5.84	23.50	4.64	1.12	5.76	0.75	4.13	0.80	1.98	1.24	0.18	1.22	1.04
2018-D	59.01	48.33	76.71	8.76	34.99	7.02	1.65	8.36	1.11	6.30	1.26	3.31	2.33	0.34	1.19	1.33
2018-E	37.98	29.82	41.67	5.17	20.21	4.09	0.97	5.16	0.68	3.93	0.78	2.04	1.36	0.20	1.22	0.93
2018-F	26.12	19.08	25.94	3.20	12.59	2.62	0.63	3.58	0.44	2.68	0.51	1.35	0.90	0.16	1.31	0.85
2018-G	45.90	30.04	40.15	5.02	20.07	3.99	1.01	5.33	0.75	4.41	0.89	2.29	1.45	0.20	1.19	0.84
2018-I	51.30	40.12	57.96	7.10	28.35	5.35	1.29	6.71	0.87	5.13	1.02	2.59	1.61	0.22	1.23	1.24
2018-J	45.77	40.11	60.44	6.96	27.93	5.35	1.24	6.53	0.84	4.95	0.98	2.54	1.69	0.26	1.22	1.18
2018-L	47.81	36.98	45.10	5.35	20.69	3.70	0.91	5.41	0.68	4.06	0.82	2.07	1.14	0.16	1.31	1.29
2018-M	45.21	36.93	46.48	5.58	21.15	3.95	0.99	5.73	0.71	4.13	0.80	1.97	1.17	0.15	1.32	1.39
2018-N	29.51	17.85	24.78	3.31	12.89	2.72	0.66	3.85	0.47	2.88	0.57	1.56	1.02	0.16	1.33	0.95
2018-O	36.41	25.02	34.48	4.42	16.85	3.28	0.80	4.56	0.57	3.44	0.69	1.90	1.28	0.17	1.30	1.06

98

99

100 Supplementary table 3. REE+Y abundances (ng/g) in limpet shells from Fuerteventura and Bay of Brest.

101

#	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Yb	Lu	Gd/Gd*	ΔGd
<i>Patella candei</i> , Fuerteventura, 2017.																
PF1	11.35	11.35	7.60	1.55	6.54	1.28	0.26	1.92	0.18	1.07	0.16	0.33	0.16	-	1.60	0.72
PF3	13.26	10.82	8.08	1.63	6.93	1.36	0.28	2.07	0.19	1.03	0.18	0.40	0.20	-	1.68	0.83
PF4	8.98	13.68	9.13	1.80	7.29	1.31	0.32	1.81	0.15	0.80	0.12	0.25	0.14	-	1.72	0.76
PF5	15.76	12.95	9.17	2.02	8.74	1.74	0.37	2.73	0.25	1.35	0.25	0.58	0.34	-	1.66	1.09
<i>Patella vulgata</i> , Bay of Brest, 2018.																
PM3	9.18	9.74	12.78	1.75	6.97	1.33	0.31	8.01	0.17	0.86	0.16	0.37	0.23		6.97	6.87
PMOUL3	19.01	16.82	22.06	2.86	11.23	2.31	0.53	15.14	0.37	1.96	0.37	0.93	0.65	0.08	6.53	12.82
PMOUL4	33.39	63.63	60.29	6.44	23.48	3.57	0.77	4.99	0.53	2.59	0.47	1.01	0.46	0.04	1.46	1.57

102

Supplementary references

- (S1) Chauvaud, L. La coquille Saint-Jacques en rade de Brest: Un modèle biologique d'étude des réponses de la faune benthique aux fluctuations de l'environnement, Ph.D. thesis, 265 pp., Univ. Bretagne Occidentale, Brest, France (1998).
- (S2) Potts, P.J., Thompson, M., Kane, J.S., Webb, P.C. and Carignan, J. GeoPT6. An international proficiency test. for analytical geochemistry laboratories - report on round 6 (OU3: Nanhoron microgranite) and 6A (CAL-S: CRPG limestone). *Geostandards Newsletter* 24 (1), E1–E37 (200