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SUPPLEMENTARY MATERIAL

Temporal variation of trace elements, rare earth elements and Pb isotope ratios in sediment core from Kiel Bay, western Baltic Sea

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Chemicals

Ultrapure water with resistivity > 18 MΩ cm was obtained from Milli-Q Element system (Millipore, USA) and was used throughout this work. Trace grade HNO_3, HF, H_2O_2 used for samples digestion were purchased from Fisher Scientific (Hampton, USA). TraceCert standard solutions for Al, Zn, Mn and Fe used for Flame AAS calibration, TraceCert Hg standard solution for AMA calibration and TraceCert multielemental standard for ICP-MS calibration, were purchased from Sigma Aldridrich (Buchs, Switzerland) in concentrations of 1000 mg L^{-1} (mono-elemental standards) and 10 mg L^{-1} (multielemental standard). Working standard solutions for REEs (containing Ce, Dy, Er, Eu, Gd, Ho, La, Lu, Nd, Pr, Sm, Sc, Tb, Tm, Yb and Y) were prepared on a daily basis by stepwise dilution of the multi-element stock standard solution (50 mg l−1) REE mix for ICP, TraceCert®, Buchs, Switzerland). MeHg standard solution, certified as 1 mg L^{-1} as mercury, was purchased from Brooks Rand (Seattle, WA, USA). The Certified Reference Materials (CRM) MESS-2 and MESS-3 (Estuarine sediments, National Research Council, Canada) were used for verification of the sample preparation method and the entire analytical procedures for TE and total Hg applied in this study. The CRMs IAEA 158 and IAEA 456 (coastal and marine sediments respectively from the International Atomic Energy Agency, Vienna, Austria) were used as quality control samples for MeHg analysis. The Standard Reference Materials SRM 981 from the National Institute of Standards and Technology (NIST, USA), with the natural isotopic composition for Pb was used for the determination of the instrumental mass discrimination during ICP-MS measurements of Pb isotopes in sediment samples. The Hg standard NIST SRM 3133 ([Hg] = 9 954 $mg.kg^{-1}$, lot No. 061204, NIST, Gaithersburg, USA) was used as a reference in the calculation of the δ values. Secondary standard UM-Almadén (NIST-8610, NIST, USA) was analyzed to assess the accuracy and the precision of the CVG/MC-ICP-MS measurements for Hg isotope ratios. HNO_3 solution at concentration of 10% v/v (Instra-Analyzed grade, 66%, JT Baker) and an HCl concentration of 2% v/v (Instra-Analyzed grade, 37%, JT Baker) were used for dilution of decomposed sediment samples before Hg isotope ratios measurements. The Tl standard NIST SRM 997 was used as internal standard for the correction of the instrumental mass bias during the Hg isotope ratios measurements. For the cold vapor generation, a reagent grade $SnCl_2$ dihydrate powder (Scharlau, Spain) was dissolved to have a final $SnCl_2$ concentration of 3% (m/m) in a 10% (v/v) HCl solution. Sample mineralization for Hg isotopic analysis were carried out using HNO_3 (Instra-Analyzed grade, 66%, JT Baker), HCl (Instra-Analyzed grade, 37%, JT Baker) and H_2O_2 (Optima grade, 30%, Fisher). All Teflon and PE lab ware used in the sample preparation and analysis, was pre-cleaned with a procedure including 12h 10% HNO_3 baths followed by the careful rinsing with MilliQ water and drying under laminar clean hood class 100.

Table S1. Trace and major elements mass fractions (in mg kg^{-1}) together with expanded uncertainty (n=3, k=2) determined in the sediment core

Depth, cm	Li	Mn	Co	Ni	Cu	Cd	Ba	Pb	U	Zn	Cr	As	Hg	Al	Fe
0-1	20 ± 3	790 ± 10	6.1 ± 0.9	17 ± 2	27 ± 4	0.62 ± 0.1	260 ± 40	41 ± 6	2.3 ± 0.3	140 ± 20	45 ± 7	13 ± 2	0.398 ± 0.06	50000 ± 7000	33000 ± 5000
1-2	25 ± 4	380 ± 60	8 ± 1	22 ± 3	38 ± 6	0.70 ± 0.1	300 ± 50	51 ± 8	3.7 ± 0.5	170 ± 30	52 ± 8	14 ± 2	0.396 ± 0.06	53000 ± 8000	34000 ± 5000
2-3	26 ± 4	350 ± 50	9 ± 1	24 ± 4	43 ± 6	0.77 ± 0.1	340 ± 50	51 ± 8	4.2 ± 0.6	160 ± 20	54 ± 8	13 ± 2	0.425 ± 0.06	50000 ± 8000	33000 ± 5000
3-4	29 ± 4	360 ± 50	10 ± 2	28 ± 4	50 ± 7	0.76 ± 0.1	390 ± 60	56 ± 8	4.1 ± 0.6	170 ± 30	57 ± 8	13 ± 2	0.558 ± 0.08	53000 ± 8000	34000 ± 5000
4-5	30 ± 4	370 ± 60	10 ± 2	29 ± 4	51 ± 8	0.81 ± 0.1	430 ± 60	55 ± 8	3.6 ± 0.5	170 ± 30	59 ± 9	12 ± 2	0.419 ± 0.06	54000 ± 8000	35000 ± 5000
5-6	27 ± 4	360 ± 50	10 ± 2	29 ± 4	48 ± 7	0.75 ± 0.1	430 ± 60	48 ± 7	3.0 ± 0.5	150 ± 20	55 ± 8	12 ± 2	0.365 ± 0.05	44000 ± 7000	32000 ± 5000
6-7	31 ± 5	360 ± 50	12 ± 2	33 ± 5	62 ± 9	1.00 ± 0.1	360 ± 50	62 ± 9	3.3 ± 0.5	190 ± 30	64 ± 10	14 ± 2	0.412 ± 0.06	43000 ± 6000	32000 ± 5000
7-8	31 ± 5	390 ± 60	12 ± 2	36 ± 5	67 ± 10	1.26 ± 0.2	420 ± 60	67 ± 10	3.7 ± 0.6	220 ± 30	69 ± 10	17 ± 2	0.445 ± 0.07	50000 ± 7000	37000 ± 5000
8-9	25 ± 4	390 ± 60	9 ± 1	26 ± 4	42 ± 6	1.18 ± 0.2	330 ± 50	60 ± 9	3.7 ± 0.6	200 ± 30	63 ± 9	14 ± 2	0.402 ± 0.06	40000 ± 6000	34000 ± 5000
9-10	26 ± 4	410 ± 60	10 ± 1	28 ± 4	46 ± 7	1.31 ± 0.2	300 ± 50	66 ± 10	3.6 ± 0.5	200 ± 30	65 ± 10	14 ± 2	0.465 ± 0.07	41000 ± 6000	34000 ± 5000
10-11	26 ± 4	380 ± 60	10 ± 1	27 ± 4	45 ± 7	1.31 ± 0.2	340 ± 50	68 ± 10	3.9 ± 0.6	200 ± 30	69 ± 10	15 ± 2	0.607 ± 0.09	38000 ± 6000	33000 ± 5000
11-12	17 ± 3	330 ± 50	6 ± 1	19 ± 3	22 ± 3	0.54 ± 0.1	350 ± 50	31 ± 5	2.0 ± 0.3	90 ± 10	40 ± 6	10 ± 1	0.434 ± 0.07	39000 ± 6000	23000 ± 3000
12-13	13 ± 2	290 ± 40	4.9 ± 0.7	16 ± 2	18 ± 3	0.45 ± 0.1	240 ± 40	24 ± 4	1.6 ± 0.2	70 ± 11	28 ± 4	7 ± 1	0.434 ± 0.07	28000 ± 4000	14000 ± 2000
13-14	16 ± 2	340 ± 50	6.3 ± 0.9	18 ± 3	25 ± 4	0.66 ± 0.1	370 ± 60	34 ± 5	2.9 ± 0.4	100 ± 20	39 ± 6	10 ± 1	0.322 ± 0.05	34000 ± 5000	21000 ± 3000
14-15	25 ± 4	460 ± 70	9 ± 1	31 ± 5	38 ± 6	1.11 ± 0.2	430 ± 60	54 ± 8	2.5 ± 0.4	170 ± 30	62 ± 9	13 ± 2	0.404 ± 0.06	49000 ± 7000	33000 ± 5000
15-16	17 ± 3	300 ± 50	7 ± 1	18 ± 3	25 ± 4	0.78 ± 0.1	430 ± 60	37 ± 6	2.0 ± 0.3	120 ± 20	39 ± 6	8 ± 1	0.271 ± 0.04	34000 ± 5000	22000 ± 3000
16-17	23 ± 3	360 ± 50	8 ± 1	23 ± 3	32 ± 5	1.42 ± 0.2	410 ± 60	58 ± 9	3.5 ± 0.5	190 ± 30	56 ± 8	12 ± 2	0.595 ± 0.09	47000 ± 7000	30000 ± 4000
17-18	23 ± 3	380 ± 60	8 ± 1	23 ± 3	36 ± 5	1.30 ± 0.2	450 ± 70	67 ± 10	3.6 ± 0.5	190 ± 30	58 ± 9	12 ± 2	0.622 ± 0.09	46000 ± 7000	30000 ± 5000
18-19	27 ± 4	370 ± 60	9 ± 1	27 ± 4	43 ± 6	1.28 ± 0.2	470 ± 71	83 ± 13	3.6 ± 0.5	210 ± 30	65 ± 10	12 ± 2	0.83 ± 0.1	50000 ± 7000	34000 ± 5000
19-20	25 ± 4	360 ± 50	9 ± 1	25 ± 4	38 ± 6	1.05 ± 0.2	450 ± 70	73 ± 11	3.2 ± 0.5	180 ± 30	56 ± 8	12 ± 2	0.81 ± 0.1	45000 ± 7000	31000 ± 5000

Table S2. Summary of EFs obtained using different background data. Approach 1: data from Geochemical Atlas of Europe (Selminen, 2005). Approach 2: shale average composition. Approach 3: data from Vallius et al., 2007 and Leipe et al., 2013.

	EF Approach 1												
Depth, cm	**Li**	**Mn**	**Co**	**Ni**	**Cu**	**Cd**	**Ba**	**Pb**	**Cr**	**As**	**Hg**	**Zn**	**U**
0-1	1.0	1.4	2.0	0.7	1	1.1	0.6	1.5	0.6	2.0	5	1.8	1.8
1-2	1.2	0.6	2.4	0.9	2	1.2	0.7	1.7	0.7	2.1	5	2.0	2.8
2-3	1.3	0.6	2.8	1.1	2	1.4	0.8	1.8	0.8	2.1	6	2.1	3.3
3-4	1.3	0.6	3.0	1.2	3	1.3	0.8	1.8	0.8	2.0	7	2.0	3.1
4-5	1.3	0.6	3.1	1.2	3	1.3	0.9	1.8	0.8	1.8	5	2.1	2.6
5-6	1.5	0.7	3.7	1.5	3	1.5	1.1	1.9	0.9	2.1	6	2.2	2.8
6-7	1.7	0.7	4.3	1.7	4	2.1	1.0	2.5	1.1	2.7	6	2.8	3.1
7-8	1.5	0.7	4.0	1.6	4	2.3	1.0	2.4	1.0	2.7	6	2.9	3.0
8-9	1.5	0.9	3.5	1.4	3	2.6	0.9	2.6	1.1	2.7	7	3.2	3.7
9-10	1.6	0.9	3.8	1.5	3	2.8	0.8	2.8	1.1	2.8	8	3.2	3.6
10-11	1.7	0.9	4.1	1.6	3	3.1	1.0	3.1	1.3	3.2	11	3.4	4.2
11-12	1.1	0.8	2.6	1.1	2	1.2	1.1	1.4	0.7	2.0	8	1.5	2.1
12-13	1.1	0.9	2.8	1.2	2	1.4	1.0	1.5	0.7	1.9	10	1.6	2.2
13-14	1.2	0.9	3.0	1.2	2	1.8	1.3	1.8	0.8	2.4	6	1.9	3.5
14-15	1.3	0.8	3.1	1.4	2	2.0	1.0	1.9	0.9	2.1	6	2.2	2.1
15-16	1.2	0.8	3.1	1.2	2	2.1	1.4	1.9	0.8	1.9	5	2.2	2.4
16-17	1.2	0.7	2.8	1.1	2	2.7	1.0	2.2	0.9	2.0	9	2.6	3.0
17-18	1.2	0.7	2.9	1.1	2	2.5	1.1	2.5	0.9	2.1	9	2.7	3.1
18-19	1.3	0.7	3.0	1.2	2	2.3	1.1	2.9	0.9	2.0	11	2.7	2.9
19-20	1.3	0.7	3.2	1.3	2	2.1	1.2	2.8	0.9	2.2	12	2.6	2.9

	EF Approach 2												
Depth, cm	**Li**	**Mn**	**Co**	**Ni**	**Cu**	**Cd**	**Ba**	**Pb**	**Cr**	**As**	**Hg**	**Zn**	**U**
0-1	0.5	1.5	0.5	0.4	1.0	3.3	0.7	3.3	0.8	1.6	1.6	2.4	1.0
1-2	0.6	0.7	0.6	0.5	1.2	3.5	0.8	3.8	0.9	1.6	1.5	2.6	1.5
2-3	0.6	0.7	0.7	0.6	1.5	4.1	0.9	4.1	1.0	1.6	1.7	2.7	1.8
3-4	0.7	0.6	0.8	0.6	1.7	3.8	1.0	4.2	0.9	1.6	2.1	2.7	1.7
4-5	0.7	0.6	0.8	0.6	1.7	4.0	1.1	4.1	1.0	1.4	1.5	2.7	1.4
5-6	0.7	0.8	1.0	0.8	2.0	4.6	1.3	4.4	1.1	1.6	1.7	2.9	1.5
6-7	0.9	0.8	1.1	0.9	2.5	6.2	1.1	5.8	1.3	2.0	1.9	3.7	1.7
7-8	0.8	0.7	1.0	0.9	2.4	6.7	1.2	5.4	1.2	2.1	1.8	3.8	1.6
8-9	0.7	0.9	0.9	0.7	1.9	7.8	1.1	5.9	1.4	2.1	2.0	4.2	2.0
9-10	0.8	0.9	1.0	0.8	2.0	8.5	1.0	6.4	1.4	2.1	2.3	4.2	1.9
10-11	0.8	0.9	1.1	0.8	2.1	9.2	1.2	7.2	1.6	2.4	3.2	4.5	2.2
11-12	0.5	0.8	0.7	0.6	1.0	3.7	1.3	3.3	0.9	1.5	2.3	2.0	1.1
12-13	0.6	1.0	0.7	0.7	1.1	4.3	1.2	3.4	0.9	1.4	3.1	2.1	1.2
13-14	0.6	0.9	0.8	0.6	1.3	5.3	1.5	4.1	1.0	1.8	1.9	2.5	1.9
14-15	0.6	0.9	0.8	0.8	1.4	6.1	1.2	4.4	1.1	1.6	1.7	2.9	1.1
15-16	0.6	0.8	0.8	0.6	1.3	6.1	1.7	4.4	1.0	1.5	1.6	2.8	1.3
16-17	0.6	0.7	0.7	0.6	1.2	8.1	1.2	5.0	1.1	1.5	3	3.5	1.6
17-18	0.6	0.8	0.8	0.6	1.4	7.6	1.4	5.8	1.1	1.6	3	3.5	1.7
18-19	0.7	0.7	0.8	0.6	1.5	6.8	1.3	6.7	1.2	1.5	3	3.5	1.6
19-20	0.7	0.7	0.8	0.7	1.5	6.2	1.4	6.5	1.1	1.7	4	3.4	1.5

	EF Approach 3									Leipe et al. 2013
Depth, cm	Co	Ni	Cu	Cd	Pb	Cr	As	Hg	Zn	Hg EF
0-1	0.4	1	0.7	3.0	1.2	0.6	0.9	18	1.0	1.20
1-2	0.5	1	1.0	3.1	1.4	0.7	1.0	17	1.1	1.11
2-3	0.6	1	1.2	3.6	1.5	0.7	1.0	19	1.2	1.27
3-4	0.6	1	1.3	3.4	1.5	0.7	0.9	24	1.1	1.58
4-5	0.6	1	1.3	3.5	1.5	0.7	0.9	17	1.2	1.16
5-6	0.8	1	1.5	4.1	1.6	0.8	1.0	19	1.2	1.25
6-7	0.9	2	1.9	5.5	2.1	1.0	1.2	22	1.6	1.44
7-8	0.8	1	1.8	6.0	1.9	0.9	1.2	20	1.6	1.35
8-9	0.7	1	1.4	6.9	2.1	1.1	1.3	22	1.8	1.49
9-10	0.8	1	1.5	7.6	2.3	1.1	1.3	26	1.8	1.70
10-11	0.9	1	1.6	8.2	2.6	1.2	1.5	36	1.9	2.41
11-12	0.6	1	0.8	3.3	1.2	0.7	0.9	25	0.8	1.70
12-13	0.6	1	0.8	3.8	1.2	0.7	0.9	35	0.9	2.33
13-14	0.6	1	1.0	4.7	1.5	0.8	1.1	22	1.1	1.44
14-15	0.6	1	1.1	5.4	1.6	0.9	1.0	19	1.3	1.25
15-16	0.6	1	1.0	5.5	1.6	0.8	0.9	18	1.2	1.19
16-17	0.6	1	0.9	7.3	1.8	0.8	0.9	29	1.5	1.92
17-18	0.6	1	1.1	6.7	2.1	0.9	1.0	31	1.5	2.04
18-19	0.6	1	1.2	6.1	2.4	0.9	0.9	38	1.5	2.51
19-20	0.7	1	1.1	5.5	2.3	0.8	1.0	40	1.5	2.69

Table S3. Geoaccumulation Indices (Igeo) calculated using the three described approaches for background evaluation.

	Igeo Approach 1												
Depth, cm	**Li**	**Mn**	**Co**	**Ni**	**Cu**	**Cd**	**Ba**	**Pb**	**Cr**	**As**	**Hg**	**Zn**	**U**
0-1	-0.3	0.2	0.7	-0.7	0.3	-0.1	-1.0	0.3	-0.9	0.7	2.1	0.6	0.6
1-2	0.0	-0.8	1.1	-0.3	0.7	0.1	-0.8	0.6	-0.7	0.9	2.1	0.8	1.3
2-3	0.1	-1.0	1.2	-0.2	0.9	0.2	-0.6	0.6	-0.7	0.8	2.2	0.8	1.5
3-4	0.2	-0.9	1.4	0.0	1.2	0.2	-0.4	0.7	-0.6	0.8	2.6	0.8	1.5
4-5	0.3	-0.9	1.5	0.1	1.2	0.3	-0.3	0.7	-0.5	0.7	2.2	0.9	1.2
5-6	0.1	-0.9	1.4	0.1	1.1	0.1	-0.3	0.5	-0.6	0.6	2.0	0.7	1.0
6-7	0.3	-0.9	1.6	0.3	1.5	0.6	-0.6	0.8	-0.4	0.9	2.2	1.0	1.1
7-8	0.3	-0.8	1.7	0.4	1.6	0.9	-0.3	1.0	-0.3	1.1	2.3	1.2	1.3
8-9	0.0	-0.8	1.3	-0.1	0.9	0.8	-0.7	0.8	-0.4	0.9	2.2	1.1	1.3
9-10	0.1	-0.7	1.4	0.0	1.0	1.0	-0.8	0.9	-0.4	0.9	2.4	1.1	1.3
10-11	0.1	-0.8	1.4	0.0	1.0	1.0	-0.6	1.0	-0.3	1.0	2.8	1.1	1.4
11-12	-0.5	-1.0	0.8	-0.5	0.0	-0.3	-0.6	-0.1	-1.1	0.4	2.3	-0.1	0.4
12-13	-0.9	-1.2	0.4	-0.8	-0.4	-0.6	-1.1	-0.5	-1.6	-0.2	2.3	-0.5	0.1
13-14	-0.6	-1.0	0.8	-0.6	0.1	0.0	-0.5	0.0	-1.1	0.4	1.8	0.1	1.0
14-15	0.0	-0.6	1.3	0.2	0.8	0.7	-0.3	0.6	-0.4	0.8	2.2	0.9	0.8
15-16	-0.5	-1.2	0.8	-0.6	0.2	0.2	-0.3	0.1	-1.1	0.1	1.6	0.3	0.4
16-17	-0.1	-0.9	1.1	-0.3	0.5	1.1	-0.4	0.7	-0.6	0.6	2.7	1.0	1.2
17-18	-0.1	-0.8	1.1	-0.2	0.7	0.9	-0.2	0.9	-0.5	0.7	2.8	1.0	1.3
18-19	0.1	-0.9	1.3	0.0	0.9	0.9	-0.2	1.3	-0.4	0.7	3.2	1.2	1.3
19-20	0.0	-0.9	1.3	-0.1	0.8	0.6	-0.2	1.1	-0.6	0.7	3.2	1.0	1.1

	Igeo Approach 2												
Depth, cm	**Li**	**Mn**	**Co**	**Ni**	**Cu**	**Cd**	**Ba**	**Pb**	**Cr**	**As**	**Hg**	**Zn**	**U**
0-1	-2.3	-0.7	-2.2	-2.6	-1.3	0.5	-1.8	0.5	-1.6	-0.6	-0.6	0.0	-1.3
1-2	-2.0	-1.8	-1.8	-2.2	-0.8	0.6	-1.5	0.8	-1.4	-0.5	-0.6	0.2	-0.6
2-3	-1.9	-1.9	-1.7	-2.1	-0.6	0.8	-1.4	0.8	-1.3	-0.5	-0.5	0.2	-0.4
3-4	-1.8	-1.8	-1.5	-1.9	-0.4	0.7	-1.2	0.9	-1.3	-0.5	-0.1	0.2	-0.4
4-5	-1.7	-1.8	-1.4	-1.8	-0.4	0.8	-1.0	0.9	-1.2	-0.7	-0.5	0.3	-0.6
5-6	-1.9	-1.8	-1.5	-1.8	-0.5	0.7	-1.0	0.7	-1.3	-0.7	-0.7	0.1	-0.9
6-7	-1.7	-1.8	-1.3	-1.6	-0.1	1.2	-1.3	1.0	-1.1	-0.4	-0.5	0.4	-0.7
7-8	-1.7	-1.7	-1.2	-1.5	0.0	1.5	-1.1	1.2	-1.0	-0.2	-0.4	0.6	-0.6
8-9	-2.0	-1.7	-1.7	-2.0	-0.7	1.4	-1.4	1.0	-1.1	-0.5	-0.6	0.5	-0.6
9-10	-1.9	-1.7	-1.5	-1.9	-0.6	1.5	-1.5	1.1	-1.1	-0.5	-0.4	0.5	-0.6
10-11	-1.9	-1.7	-1.6	-1.9	-0.6	1.5	-1.4	1.2	-1.0	-0.4	0.0	0.5	-0.5
11-12	-2.5	-1.9	-2.2	-2.5	-1.6	0.3	-1.3	0.1	-1.8	-1.0	-0.5	-0.7	-1.4
12-13	-2.9	-2.1	-2.5	-2.7	-1.9	0.0	-1.9	-0.3	-2.3	-1.6	-0.5	-1.1	-1.8
13-14	-2.6	-1.9	-2.2	-2.5	-1.5	0.6	-1.2	0.2	-1.8	-1.0	-0.9	-0.5	-0.9
14-15	-2.0	-1.5	-1.6	-1.7	-0.8	1.3	-1.0	0.8	-1.1	-0.6	-0.6	0.3	-1.1
15-16	-2.5	-2.1	-2.1	-2.5	-1.4	0.8	-1.0	0.3	-1.8	-1.3	-1.1	-0.3	-1.4
16-17	-2.1	-1.8	-1.8	-2.2	-1.1	1.7	-1.1	0.9	-1.3	-0.7	0.0	0.4	-0.7
17-18	-2.1	-1.7	-1.8	-2.1	-0.9	1.5	-0.9	1.1	-1.2	-0.7	0.1	0.4	-0.6
18-19	-1.9	-1.8	-1.6	-1.9	-0.7	1.5	-0.9	1.5	-1.1	-0.7	0.5	0.6	-0.6
19-20	-2.0	-1.8	-1.7	-2.0	-0.8	1.2	-0.9	1.3	-1.3	-0.6	0.4	0.4	-0.8

	Igeo Approach 3								
Depth, cm	Co	Ni	Cu	Cd	Pb	Cr	As	Hg	Zn
0-1	-1.7	-1.0	-0.9	1.1	-0.2	-1.1	-0.5	3.7	-0.4
1-2	-1.3	-0.6	-0.4	1.3	0.1	-0.9	-0.4	3.7	-0.2
2-3	-1.2	-0.5	-0.2	1.4	0.1	-0.9	-0.4	3.8	-0.2
3-4	-1.0	-0.3	0.0	1.4	0.2	-0.8	-0.4	4.2	-0.2
4-5	-1.0	-0.2	0.0	1.5	0.2	-0.8	-0.5	3.8	-0.1
5-6	-1.0	-0.2	0.0	1.4	0.0	-0.9	-0.6	3.6	-0.3
6-7	-0.8	0.0	0.3	1.8	0.4	-0.7	-0.3	3.8	0.0
7-8	-0.7	0.1	0.4	2.1	0.5	-0.5	-0.1	3.9	0.3
8-9	-1.2	-0.4	-0.2	2.1	0.3	-0.7	-0.4	3.7	0.1
9-10	-1.1	-0.3	-0.1	2.2	0.5	-0.6	-0.4	4.0	0.1
10-11	-1.1	-0.3	-0.1	2.2	0.5	-0.5	-0.3	4.3	0.1
11-12	-1.7	-0.8	-1.2	0.9	-0.6	-1.3	-0.9	3.9	-1.0
12-13	-2.0	-1.1	-1.5	0.7	-1.0	-1.9	-1.5	3.9	-1.4
13-14	-1.7	-0.9	-1.0	1.2	-0.5	-1.4	-0.9	3.4	-0.9
14-15	-1.1	-0.1	-0.4	2.0	0.2	-0.7	-0.5	3.8	-0.1
15-16	-1.6	-0.9	-1.0	1.5	-0.3	-1.4	-1.1	3.2	-0.7
16-17	-1.3	-0.6	-0.6	2.3	0.3	-0.8	-0.6	4.3	0.0
17-18	-1.3	-0.5	-0.5	2.2	0.5	-0.8	-0.6	4.4	0.0
18-19	-1.1	-0.3	-0.2	2.2	0.8	-0.6	-0.6	4.8	0.2
19-20	-1.2	-0.4	-0.4	1.9	0.6	-0.8	-0.5	4.7	0.0

Leipe et al., 2013
Hg
-0.2
-0.2
-0.1
0.3
-0.1
-0.3
-0.1
0.0
-0.2
0.0
0.4
-0.1
-0.1
-0.5
-0.2
-0.7
0.4
0.5
0.9
0.8

Table S4. Hg isotopic composition expressed in ‰. Uncertainty (2SD) was calculated by multiple measurements (n=9) of UM-Almaden (NIST 8610) secondary standard

Depth, cm	**δ^{204}Hg**	**δ^{202}Hg**	**δ^{201}Hg**	**δ^{200}Hg**	**δ^{199}Hg**	**Δ^{204}Hg**	**Δ^{201}Hg**	**Δ^{200}Hg**	**Δ^{199}Hg**
0-1	-0.97	-0.59	-0.46	-0.31	-0.13	-0.09	-0.02	-0.02	0.02
1-2	-0.80	-0.49	-0.33	-0.19	-0.04	-0.07	0.04	0.06	0.08
10-11	-0.88	-0.54	-0.39	-0.25	-0.09	-0.08	0.01	0.02	0.05
16-17	-0.94	-0.58	-0.48	-0.27	-0.16	-0.07	-0.05	0.02	-0.02
17-18	-0.85	-0.60	-0.53	-0.31	-0.19	0.04	-0.08	-0.01	-0.04
19-20	-0.95	-0.61	-0.44	-0.33	-0.15	-0.03	0.02	-0.02	0.00
Average	-0.90	-0.57	-0.44	-0.28	-0.13	-0.05	-0.01	0.01	0.02
Uncertainty	*0.18*	*0.12*	*0.09*	*0.08*	*0.06*	*0.11*	*0.02*	*0.07*	*0.05*

Table S5. Pb isotope ratios results presented together with expanded uncertainty (k=2)

Depth, cm	$^{206}Pb/^{204}Pb$			$^{207}Pb/^{204}Pb$			$^{208}Pb/^{204}Pb$			$^{208}Pb/^{206}Pb$			$^{206}Pb/^{207}Pb$		
0-1	18.65	±	0.08	15.72	±	0.05	38.5	±	0.1	2.067	±	0.002	1.186	±	0.001
1-2	18.65	±	0.08	15.74	±	0.05	38.5	±	0.1	2.066	±	0.002	1.185	±	0.001
2-3	18.51	±	0.08	15.64	±	0.05	38.2	±	0.1	2.066	±	0.002	1.184	±	0.001
3-4	18.56	±	0.08	15.67	±	0.05	38.4	±	0.1	2.069	±	0.002	1.185	±	0.001
4-5	18.68	±	0.08	15.77	±	0.05	38.7	±	0.1	2.073	±	0.001	1.184	±	0.001
5-6	18.56	±	0.08	15.71	±	0.05	38.5	±	0.1	2.073	±	0.001	1.181	±	0.001
6-7	18.55	±	0.08	15.69	±	0.05	38.5	±	0.1	2.073	±	0.002	1.182	±	0.001
7-8	18.57	±	0.08	15.75	±	0.05	38.6	±	0.1	2.084	±	0.008	1.179	±	0.001
8-9	18.34	±	0.08	15.60	±	0.05	38.2	±	0.1	2.078	±	0.002	1.176	±	0.001
9-10	18.42	±	0.08	15.70	±	0.05	38.4	±	0.1	2.084	±	0.003	1.173	±	0.001
10-11	18.42	±	0.08	15.73	±	0.05	38.4	±	0.1	2.084	±	0.002	1.171	±	0.001
11-12	18.36	±	0.08	15.56	±	0.05	37.9	±	0.1	2.066	±	0.002	1.179	±	0.001
12-13	18.44	±	0.08	15.63	±	0.05	38.2	±	0.1	2.066	±	0.003	1.180	±	0.001
13-14	18.26	±	0.08	15.48	±	0.05	37.9	±	0.1	2.071	±	0.002	1.180	±	0.002
14-15	18.47	±	0.08	15.70	±	0.05	38.3	±	0.1	2.074	±	0.002	1.177	±	0.001
15-16	18.51	±	0.08	15.72	±	0.05	38.6	±	0.2	2.086	±	0.003	1.177	±	0.001
16-17	18.31	±	0.08	15.55	±	0.05	38.1	±	0.1	2.078	±	0.002	1.178	±	0.002
17-18	18.48	±	0.08	15.72	±	0.05	38.4	±	0.1	2.082	±	0.002	1.176	±	0.001
18-19	18.54	±	0.08	15.76	±	0.05	38.6	±	0.1	2.082	±	0.002	1.177	±	0.001
19-20	18.32	±	0.08	15.59	±	0.05	38.1	±	0.1	2.079	±	0.002	1.175	±	0.001

Fig. S1. Map showing the sampling station in Kiel bay (black star)