Metals and organotins in multiple bivalve species in a one-off global survey

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7 figures (SI-1 to SI-7)

6 tables (SI-1 to SI-6)

Correlation of metals and TBT

A simple correlation test (Microsoft Excel 2003, Data analysis toolkit, CORREL function) of the *Mytilus*-normalised data indicates that only for TBT, DBT and MBT, concentrations are strongly correlated, with correlation coefficients of 0.85 to 0.89 (table SI-4), also visible in the PCA loading plot (figure SI-3). Fe and Ni concentrations also correlate with a coefficient of 0.89; otherwise most concentration correlations are below 0.5. Concentrations of Cr in mussel tissue seem correlated with those of Ca and Al, with coefficients of 0.74 and 0.60 respectively. An interesting observation is that Cu concentrations correlate relatively strongly with DBT concentrations at 0.73, but also with TBT and MBT concentrations at 0.49 respectively 0.47. Cu is a main component of antifouling paints, also those containing TBT, typically 10-30% (up to 50%) of the paint is Cu_2O or CuSCN. DBT is the first debutylation step of breakdown from TBT to Sn.

Assessment criteria for Cu and Zn

The OSPAR QSR 2010 did not assess Cu and Zn, so no revised LC and BAC are available. Instead, data are compared to the older Background/Reference Concentrations (B/RC) revised in 2005[1]. The B/RCs was a forerunner for LC and BAC, and is set up as a range, for Cu 0.76 to 1.11 mg kg^{-1} WW and for Zn 11.6 to 30 mg kg^{-1} WW, setting a range for the expected reference concentration for metals in the North Sea. A default conversion factor of 20% DW is used to convert B/RC values from WW to DW. The upper B/RC is lower than the LC used in the QSR 2010 (table 1) for Cd, Hg and Pb, at 56-73% of the LC used for these metals. Both Cu and Zn are essential elements actively taken up by mussels, and no EAC or EC food limit are set for these metals.

Assessment of Cu

Concentration of Cu was high in the four *Isognomen* and *Brachidontes* samples from US Virgin Islands (figure Si-1). In the PCA analysis, they were clustered around approx. (1.0, 0.5) after species correction (figure SI-6), indicating a small residual Cu anomaly after normalisation. The species from the *Mytilidae* family at the US Virgin Islands (*Brachidontes)* was measured at twice the highest Cu concentration found at Perth industrial harbour. This could either indicate that there are intra-family differences for Cu between the *Mytilus* and *Brachidontes* Genus, that also needs to be taken into account when establishing the family correction factors or that Cu concentrations in mangroves generally are very high. The highest concentrations are found around the Solomon Islands (Rar1_GEL_R), New Zealand (NZ3_MYT_M) and Perth (Per7_MYT_M). At the other end of the scale, 15% of the samples were below the upper B/RC, with three sites below the lower B/RC: the reference station for Sydney (Syd1_SAC_R), Solomon Islands (Rar3_SAC_H), and *Saccostrea* and *Mytilus* from Hobart (Tas2_SAC_M; Tas2_MYT_M). Most samples from the Southern hemisphere were below the upper B/RC. From the Northern Hemisphere only two *Chlamys* reference stations from Greenland (GRL_CHL_R) and Faroe Islands (Rar1_CHL_R) were below the upper B/RC. Concentration differences spanned three orders of magnitude (two if the US Virgin Islands is disregarding), and the median concentration was two times the upper B/RC value.

Assessment of Zn

For Zn, the highest concentration was found in Perth (PER7_MYT_H) at 3 times the upper B/RC concentration. There where very large correction for *Isognomon* and *Saccostrea* employed, but the normalised concentrations were within the range of *Mytilus*. Concentrations in the Open water *Pectinadea* were below the lower B/RC value after normalisation, but even before normalisation held the lowest Zn concentration. 39% of the samples were within the B/RC range, including most samples from the Greenlandic reference coast and US Virgin Islands. Concentration differences spanned two orders of magnitude for Zn, and the median concentration was within the B/RC range.

FIGURES.

Figure SI-1: Assessment of measured copper and zinc concentrations. The vertical lines indicate the different locations, with sites ordered left to right from expected reference to highly polluted sites. Note logarithmic axis. Zn concentrations are shown in blue squares and Cu in green circles, with *Mytilus* shown as open squares/circles, and *Saccostrea/Isognomon* with filled square/circle, for Zn also *Chlamys* in light blue colour. Background reference concentrations indicated (lower B/RC limit dashed – upper full). The displayed concentrations are after normalisation (factors given in table 3), *Isognomon* only found at site 12, *Saccostrea* at site 7 to 9.

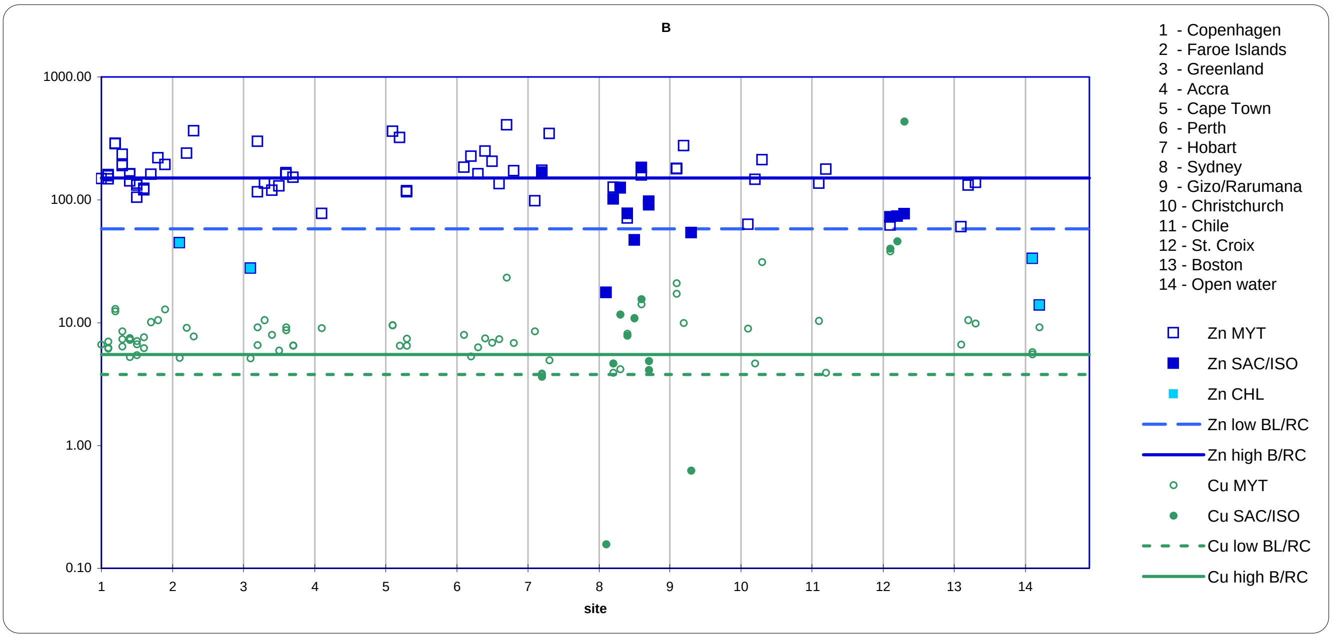


Figure SI-2: Linear relationships between *Saccostrea* and *Mytilus* from the same sites (units mg kg^{-1}). See table 2 for correlation coefficients.

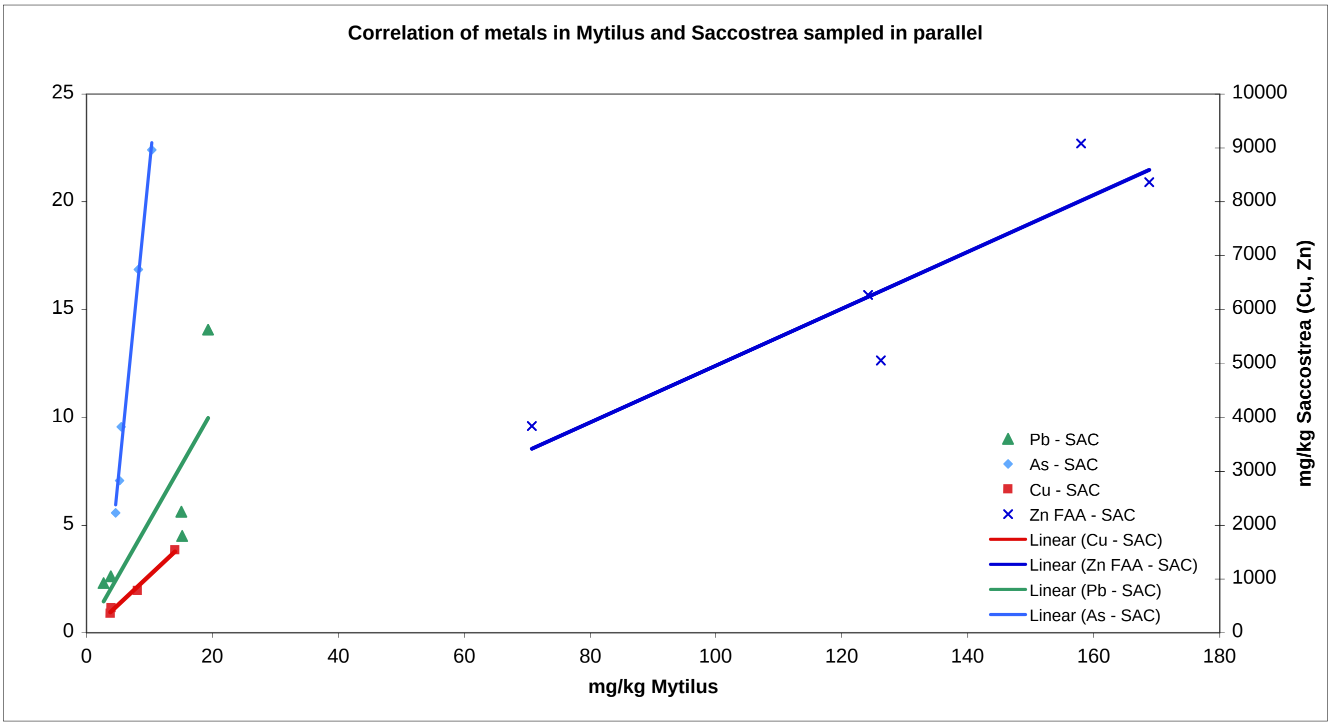


Figure SI-3: Relationships between TBT concentration (as µg TBT-Sn kg^{-1}), species (*Geloina* and *Isognomon* pooled as GEL; *Mytilus*, *Perna* and *Brachidontes* pooled as Myt) and distance to the nearest (larger) harbour. Notice logarithmic Y-scale. Lines indicate the different assessment criteria from table 1 in the main text.

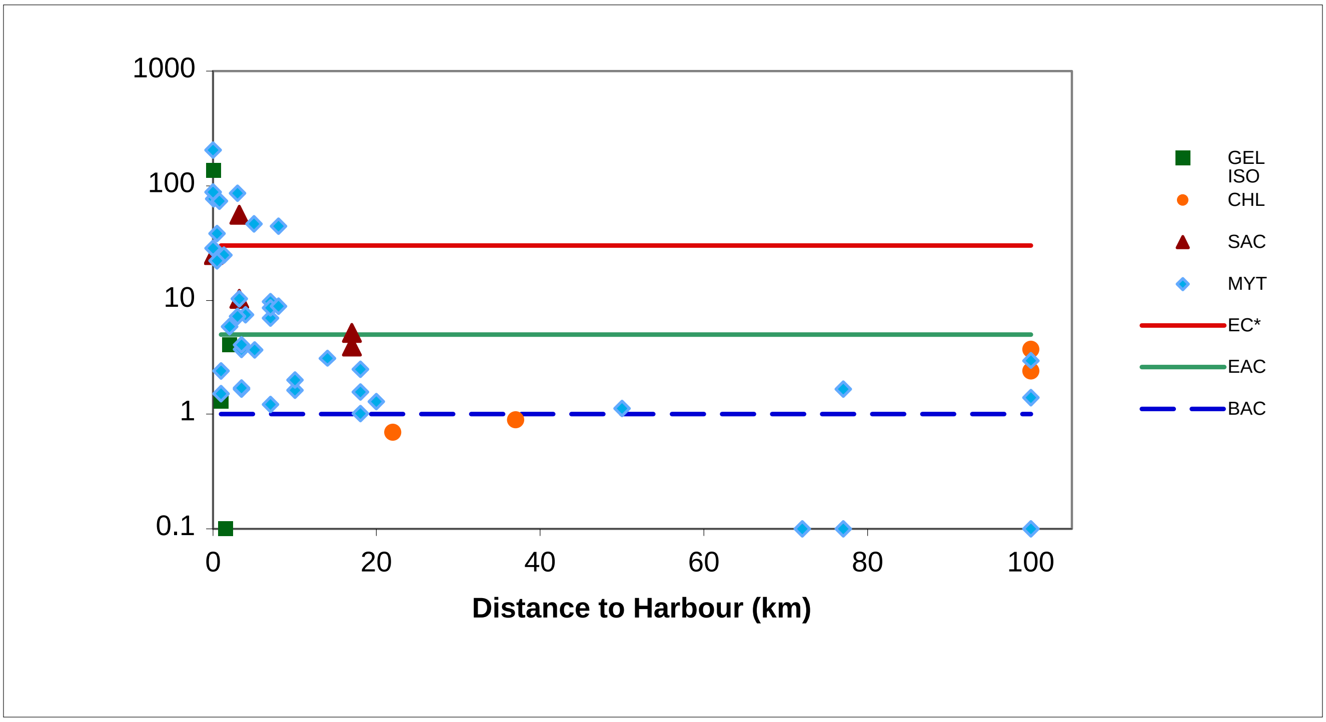


Figure SI-4: Mean centred score and loading plot for PCA analysis PC1 vs. PC2 of auto scaled concentration data before normalisation. Notice all *Saccostrea* stations (_SAC_) are in the lower left corner, separated from the bulk of the mussel data.

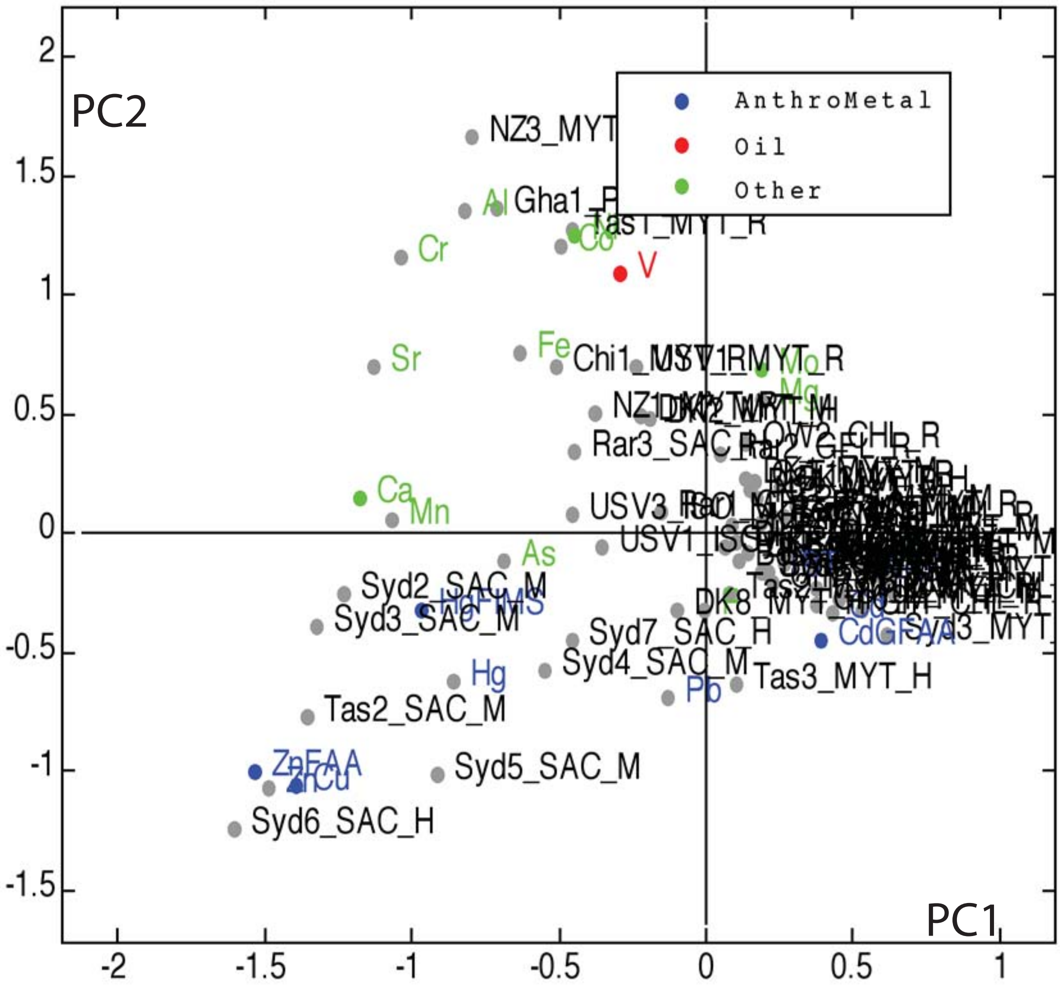


Figure SI-5: Mean centred score and loading plot for PCA analysis PC3 vs. PC4 of auto scaled concentration data before normalisation. Notice most *Chlamys* stations (_CHL_) are in the lower right corner, separated from the bulk of the mussel data.

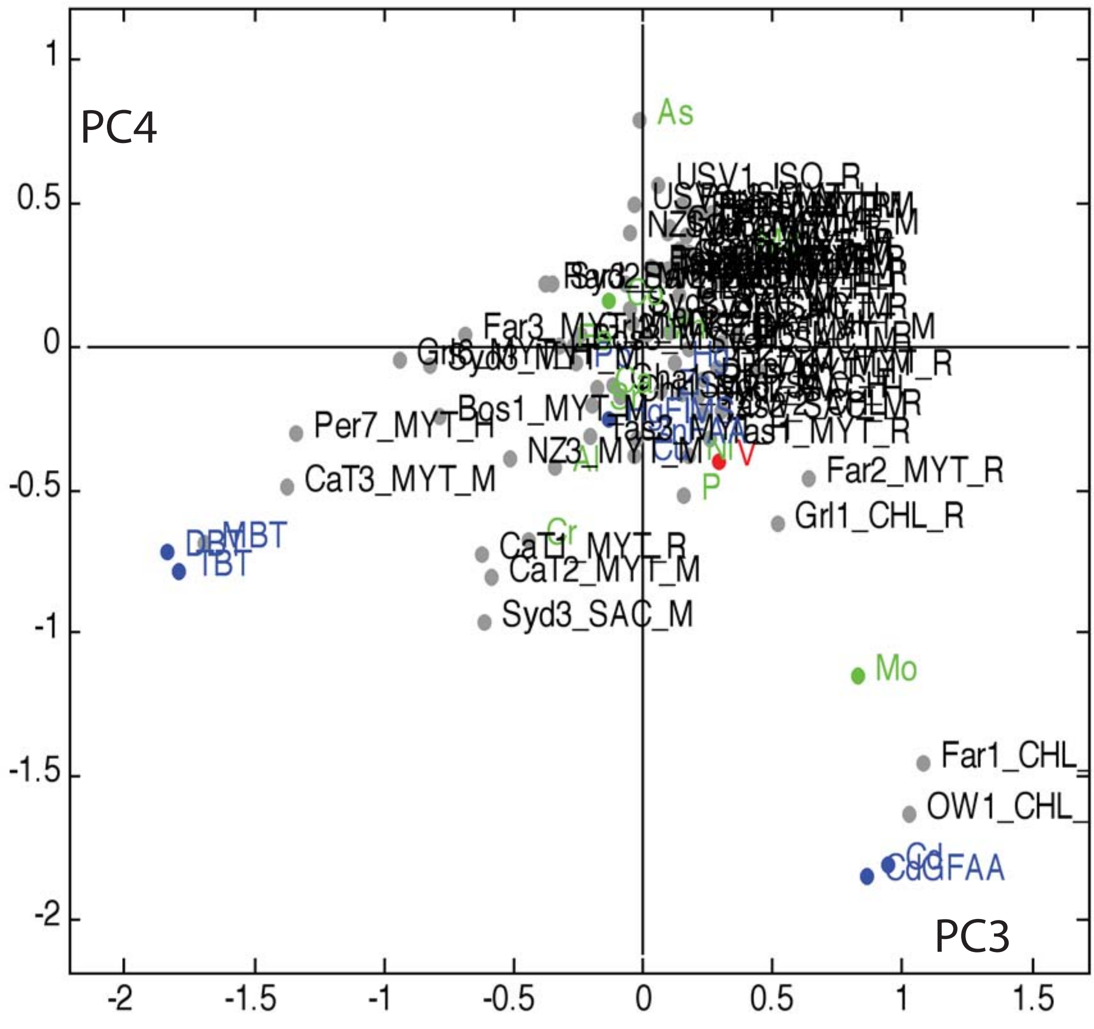


Figure SI-6. Bi-plot of PC1 (19,5%) vs. PC2 (14,8%) for PCA analysis after species normalisation to *Mytilus*, with the loadings coloured according to interpretations: Blue for Anthropogenic metals, red for oil-indicator (V), and green for other metals.

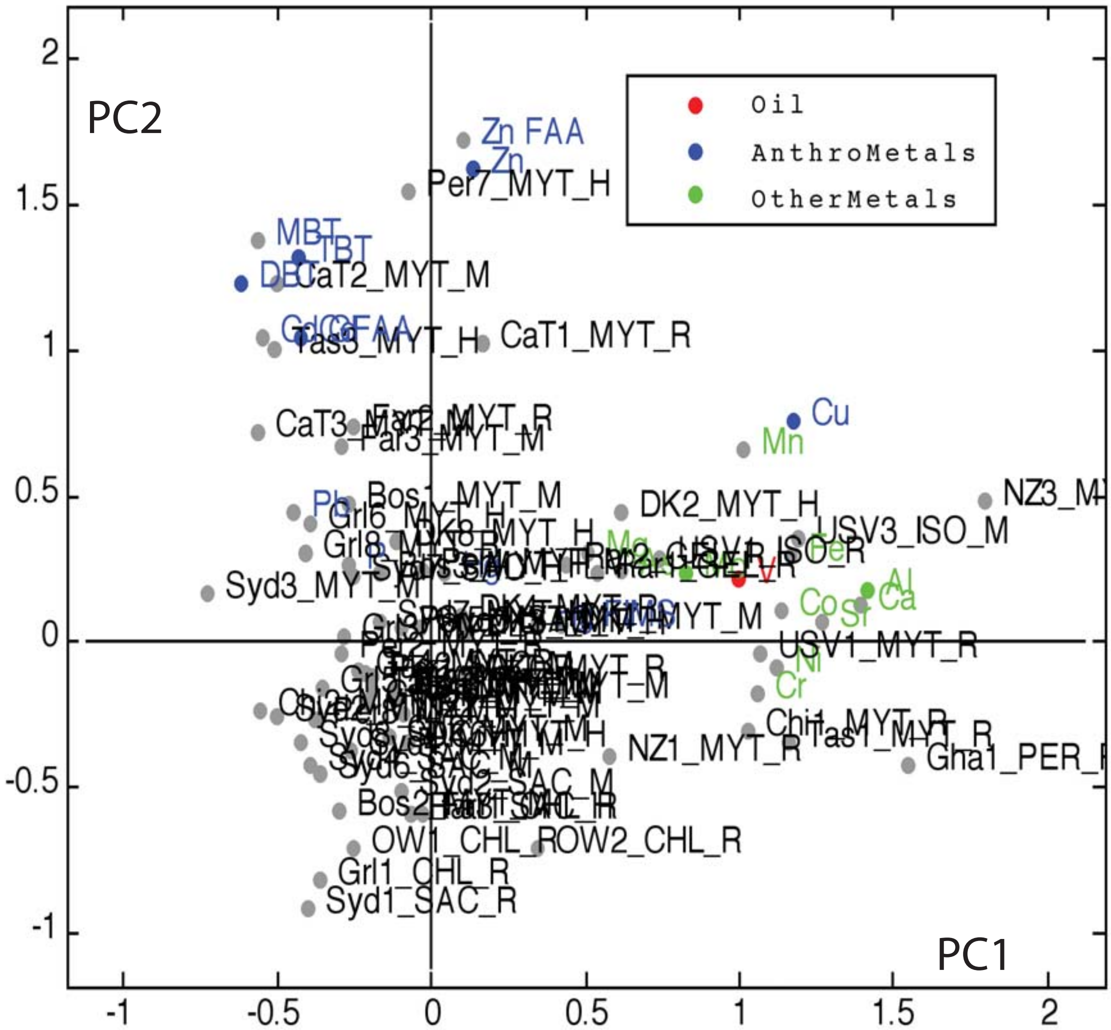
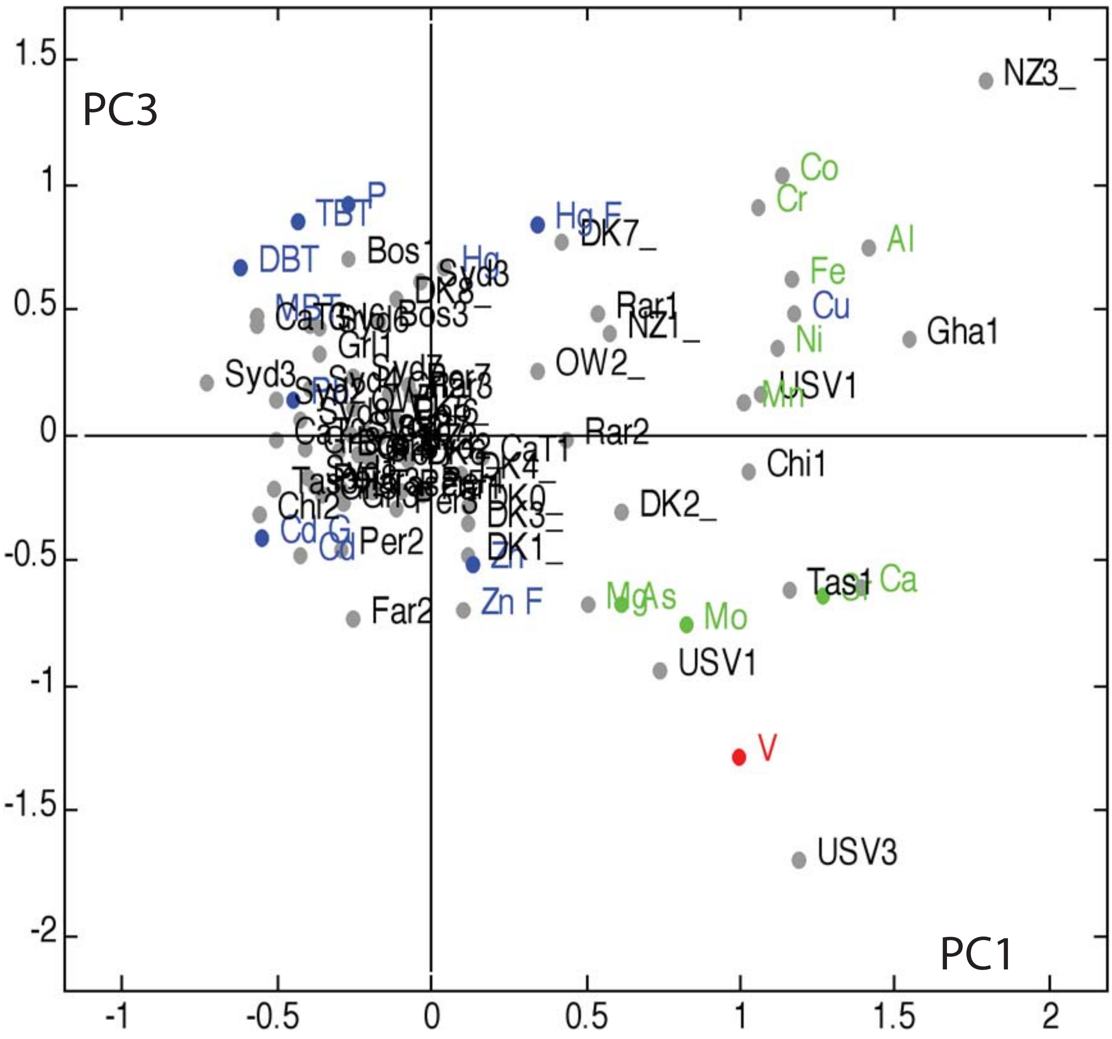


Figure SI-7. Bi-plot of PC1 (19,5%) vs. PC3 (9.3%) for PCA analysis after species normalisation to *Mytilus*, with the loadings coloured according to interpretations: Blue for Anthropogenic metals, red for oil-indicator (V), and green for other metals. The naming has been reduced to the four first letters to reduce cluttering.



TABLES.

Table SI-1. QA information for samples: Detection limits and certified reference materials (µg-Sn or mg per kg dry weigth (n=4 for NIST 2976, n=5 for CRM 477 ; n=9 for internal reference material Marina)

Element	CRM id	CRM target	Measured	RSD	DL*
Zn/FAA	NIST 2976	137	136	1.9%	7.5 mg kg^{-1}
Zn	Marina	129	121	2.3%	7.5 mg kg^{-1}
Hg/CVAA	NIST 2976	0.061	0.064	11.8%	0.02 mg kg^{-1}
Hg	Marina	0.156	0.163	2.2%	0.02 mg kg^{-1}
Cd/GFAA	NIST 2976	0.82	0.85	2.6%	0.02 mg kg^{-1}
Cd	Marina	0.72	0.98	6.3%	0.02 mg kg^{-1}
Na	NIST 2976	35000	37300	18.9%	300 mg kg^{-1}
Na	Marina	n.a.	8800	2.0%	300 mg kg^{-1}
Mg	NIST 2976	5300	5280	13.9%	50 mg kg^{-1}
Mg	Marina	n.a.	1650	2.1%	50 mg kg^{-1}
Al	NIST 2976	134	120	21%	20 mg kg^{-1}
Al	Marina	n.a.	500	11.5%	20 mg kg^{-1}
K	NIST 2976	9700	11900	13.9%	150 mg kg^{-1}
K	Marina	n.a.	4900	3.2%	150 mg kg^{-1}
Ca	NIST 2976	7600	6450	27%	250 mg kg^{-1}
Ca	Marina	n.a.	3650	8.6%	250 mg kg^{-1}
Cr	NIST 2976	0.5	0.8	35%	0.3 mg kg^{-1}
Cr	Marina	0.95	1,06	6.9%	0.3 mg kg^{-1}
Mn	NIST 2976	33	31	36%	20 mg kg^{-1}
Mn	Marina	n.a.	43	10.2	20 mg kg^{-1}
Fe	NIST 2976	171	52	250%	15 mg kg^{-1}
Fe	Marina	n.a.	492	18.8%	15 mg kg^{-1}
Ni	NIST 2976	0.93	0.87	37%	0.3 mg kg^{-1}
Ni	Marina	2.39	2.2	7.7%	0.3 mg kg^{-1}
Co	NIST 2976	0.61	<dl	49%	0.6 mg kg^{-1}
Co	Marina	n.a.	0.7	22%	0.6 mg kg^{-1}
Cu	NIST 2976	4.02	3.7	33%	2.5 mg kg^{-1}
Cu	Marina	7.01	6.4	5.4%	2.5 mg kg^{-1}
As	NIST 2976	13.3	12,6	2.5%	0.2 mg kg^{-1}
As	Marina	7.03	7,70	4.2%	0.2 mg kg^{-1}
Sr	NIST 2976	93	63	16%	1 mg kg^{-1}
Sr	Marina	n.a.	37	4.8%	1 mg kg^{-1}
Pb	NIST 2976	1.19	1.00	37%	0.3 mg kg^{-1}
Pb	Marina	1.35	0.94	11.5%	0.3 mg kg^{-1}
U	NIST 2976	0.0013	<dl	26%	0.2 mg kg^{-1}
TBT	CRM 477	900	867	12.1%	1 µg-Sn kg^{-1}
DBT	CRM 477	785	793	13.1%	1 µg-Sn kg^{-1}
MBT	CRM 477	913	855	17.4%	1 µg-Sn kg^{-1}

n.a.: No target value available; Red colour: QA data not <15% RSD, or outside of 85-115% recovery, often due to reference material close to accredited detection limit. In some cases better DL is achieved in the actual run, based on replicate analysis of blanks. In this case, concentrations above the actual DL have been reported.

Table SI-2. Bivalve species sampled, area of origin, description of the study area and distance to probable sources of heavy metals and organotins. The sample ID gives abbreviations of the site (DK# for Denmark, family code (specific species is given in bivalve species column) and the sample category (H: harbour, C: coast, R: reference). The nearest contamination source for coast/reference sites in remote location is taken as the nearest build-up area.

PCA code	Station name, comment	Area type	Bivalve species	WGS84 N	WGS 84 E	Major city/ harbour (km)	Nearest contam. source (km)	# in pool	Weighted length (mm)	Size class of mussel pool
DK0_MYT_R	Køge, coast south of Copenhagen	Reference	*Mytilus edulis*	55º29'36"N	12º25'24"E	14	12	77	44.6	40-65
DK1_MYT_M	Great Belt sea, coast of Agersø Island,	Coastal with ship traffic	*Mytilus edulis*	55º14'42"N	11º11'42"E	7	5.4	30	60.5	50-70
DK1_MYT_M	Replicate	"	"	"	"	7	5.4	25	61.3	50-80
DK1_MYT_M	Replicate	"	"	"	"	7	5.4	30	63.0	55-70
DK2_MYT_H	Roskilde Fjord, former industrial site (steel mill)	Fjord with ship traffic	*Mytilus edulis*	55º57'30"N	12º01'0"E	1	0.2	80	49.9	40-60
DK2_MYT_H	Replicate	"	"	"	"	1	0.2	80	49.9	40-60

DK3_MYT_H	Odense Fjord	Fjord with ship traffic and major Shipyard	*Mytilus edulis*	55º28'59"N	10º30'36"E	7	2	20	63.8	50-85
DK3_MYT_H	"	"	"	"	"	7	2	22	57.0	50-70
DK3_MYT_H	Analytical replicate		*Mytilus edulis*	55º28'59"N	10º30'36"E	7	2	22	57.0	50-70
DK4_MYT_R	Nivå Bay, North of Copenhagen,	Coastal with ship traffic	*Mytilus edulis*	55º54'48"N	12º32'06"E	18	1.1	31	59.0	50-70
DK4_MYT_R	Analytical replicate	"	"	"	"	18	1.1	31	59.0	50-70
DK4_MYT_R	Replicate	"	"	"	"	18	1.1	26	64.8	55-80
DK4_MYT_R	Replicate	"	"	"	"	18	1.1	28	63.9	55-75
DK5_MYT_M	Skovshoved	Coastal with ship traffic	*Mytilus edulis*	55º48'10"N	12º35'39"E	3.5	4	30	59.3	45-70
DK5_MYT_M	Analytical replicate	"	"	"	"	3.5	4	30	59.3	45-70
DK5_MYT_M	Replicate	"	"	"	"	3.5	4	24	62.9	55-70

DK6_MYT_H	Copenhagen Lynetten The sound	Close to outlet from municipal wastewater treatment plant	*Mytilus edulis*	55º43'35"N	12º37'52"E	3.5	3	33	61.9	55-70
DK6_MYT_H	replicate	"	"	"	"	3.5	3	30	62.8	55-70
DK7_MYT_M	Copenhagen harbour,	Outer pier, Trekroner fortres	*Mytilus edulis*	55º42'15"N	12º36'43"E	1.5	3	212	38.8	25-65
DK8_MYT_H	Copenhagen southern harbour	Less trafficated Harbour	*Mytilus edulis*	55º39'53"N	12º34'01"E	0.5	3	80	40.7	25-60
DK9_MYT_H	Copenhagen main harbour	Main harbour	*Mytilus edulis*	55º41'36"N	12º36'0"E	0	0.5	85	41.3	25-55
Far1_CHL_R	Open water off Nolsoy	Reference	*Chlamys opercularis*	62º02'17"N	6º30'0"W	37.0	37	20	64.8	50-75
Far2_MYT_R	Faroe Islands	Coastal reference	*Mytilus edulis*	62º01'0"N	6º45'0"W	1.5	1.5	n.r.	n.r.	n.r.
Far3_MYT_M	Skalafjord coast with ship traffic	Coastal with ship traffic	*Mytilus edulis*	62º10'54"N	6º47'56"W	1.3	1.3	n.r.	n.r.	n.r.
Grl1_CHL_R	Coast near Sisimiut	Coastal reference	*Chlamys islandica*	66º55'24"N	54º11'45"W	22.0	22	40	65.8	30- >90

Grl2_MYT_M	Central West coast near Qeqertarsuaq	Coastal with ship traffic	*Mytilus edulis*	64º27'26"N	50º14’07"W	77.0	3.0	87	54.9	45-75
Grl3_MYT_R	Bredefjord, coast	Coastal reference	*Mytilus edulis*	60º43’07"N	46º59'17"W	100.0	12.0	90	50.1	25-80
Grl4_MYT_R	Coast near Sisimiut	Coastal reference	*Mytilus edulis*	66º53'35"N	53º15'0"W	20.0	20	78	48.8	20-90
Grl5_MYT_R	Coast near Naatalik	Coastal reference	*Mytilus edulis*	60º07'18"N	45º15'16"W	100.0	2.6	20	46.0	30-55
Grl6_MYT_R	Coast near Sisimiut	Coastal reference	*Mytilus edulis*	66º56'03"N	53º44'57"E	3.0	3	49	51.8	20->90
Grl7_MYT_H	Nuuk harbour	Harbour/ marina	*Mytilus edulis*	64º10'09"N	51º43'38"W	5.1	5	99	51.7	30-85
Grl8_MYT_M	Nuuk harbour, outer pier	Outer Pier	*Mytilus edulis*	64º09'48"N	51º44'16"E	0.5	0.5	121	55.1	35-80
Gha1_PER_R	Coast near Accra	Coastal reference	*Perna perna*	5º29'17"N	0º22'30"W	20	20	152	17.3	10-35
CaT1_MYT_R	Coast near Cape Town, ship traffic	Coastal reference	*Perna perna*	33º56'38"S	18º22'24"E	8.0	1.0	152	29.8	25-50
CaT2_MYT_M	Coast near Cape Town	Coastal with ship traffic	*Perna perna*	33º54'54"S	18º23'13"E	5.0	0.5	71	41.7	30-65

CaT3_MYT_M	Pier at Cape Town	Outer Pier	*Mytilus sp*	33º53'54"S	18º25'46"E	0.8	0.8	47	77.9	55->90
Per1_MYT_R	Fremantle reference (0-1 m)	Coastal reference	*Mytilus edulis*	32º03'19"S	115º43’01’’ E	2.0	2.0	86	47.8	35-75
Per2_MYT_R	Fremantle outside harbour (0-1m)	Coastal with ship traffic	*Mytilus edulis*	32º03'14"S	115º42'24"E	3.0	3.0	66	45.7	35-65
Per3_MYT_M	Perth, Swan River (0-1 m)	River with ship traffic	*Mytilus edulis*	32º02'28"S	115º45'23"E	1.2	1.2	48	65.3	50-80
Per4_MYT_M	Perth, Swan River, The Coombe	River with ship traffic	*Mytilus edulis*	32º01’07"S	115º46'35"E	5	1.1	80	62.8	45-85
Per5_MYT_M	Perth, Swan River Freemantle Bridge (0-1m)	River with ship traffic	*Mytilus edulis*	32º00'24"S	115º47’08" E	6.0	1.4	56	45.7	35-60
Per6_MYT_M	Perth, Swan River Kingspark boathouse (0-1m)	River with ship traffic	*Mytilus edulis*	31º58'14"S	115º49'21"E	3.4	1.1	86	51.1	40-70
Per7_MYT_H	Freemantle harbour (0-1m)	Harbour	*Mytilus edulis*	32º03'34"S	115º44'11"E	0.0	0.0	119	52.0	35-70

Per8_MYT_H	Freemantle inner harbour (0-1m)	Harbour	*Mytilus edulis*	32º02'31"S	115º43'33"E	0.5	0.5	89	52.7	20-90
Tas1_MYT_R	Hobart 2	Coastal reference	*Mytilus sp*	43º01'32"S	147º55'40"E	50.0	2.0	109	36.5	30-50
Tas2_MYT_M	Hobart 1	Costal with ship traffic	*Mytilus sp*	43º01'41"S	147º24'48"E	17.0	0.5	80	47.1	35-80
Tas2_SAC_M	Species replicat	”	*Saccostrea sp*	”	”	17.0	0.5	15	50.0	n.a.
Tas3_MYT_H	Hobart 3	Harbour	*Perna perna*	42º53'05"S	147º20'0"E	0.1	1	0	50.0	40-85
Syd1_SAC_R	Lake Conjola 2	Coastal reference	*Saccostrea glomerata*	35º15'34"S	150º29'14"E	100.0	1.0	22	49.3	35-65
Syd2_SAC_M	Sydney 7 Rose Bay	Outer Pier	*Saccostrea glomerata*	33º51’02’’S	151º16'21"E	8.0	0.2	19	51.4	35-60
Syd2_MYT_M	Species replicate	”	*Mytilus sp*	”	”	8.0	0.2	34	69.3	45-90
Syd3_MYT_M	Sydney 2 Iron Cove	Outer Pier	*Mytilus sp*	33º51'56"S	151º09'18"E	3.2	0.1	0	50.0	55->90
Syd3_SAC_M	Species replicate	”	*Saccostrea glomerata*	”	”	3.2	0.1	35	63.9	40-85
Syd4_MYT_M	Sydney 4 Lane Cove	Estuarie heavy ship traffic	*Mytilus edulis*	33º49'48"S	151º08'51"E	6.0	0.1	45	70.9	40->90

Syd4_SAC_M	Species replicate	”	*Saccostrea glomerata*	”	”	6.0	0.1	24	62.1	40-80
Syd5_SAC_M	Sydney 6 Paradise Island Sydney Harbour	Outer Pier	*Saccostrea glomerata*	33º51'34"S	151º15'55"E	7.0	0.4	37	56.7	35-80
Syd6_MYT_H	Sydney 3 Darling Harbour	Harbour	*Mytilus edulis*	33º52'18"S	151º11'23"E	1.4	0.3	49	62.5	40-85
Syd6_SAC_H	Species replicate	”	*Saccostrea glomerata*	”	”	1.4	0.3	26	56.0	35-80
Syd7_SAC_H	Sydney 1 Iron Cove	Harbour	*Saccostrea glomerata*	33º52'10"S	151º8'38"E	4.2	0.1	30	70.1	40->90
Syd7_SAC_H	Analytical replicate	”	”	”	”	4.2	0.1	30	70.1	40->90
Rar1_GEL_R	Rarumana 1	Coastal reference (mangrove)	*Geloina coaxans*	8º13'58"S	157º03'21"E	27	27	0	50.0	
Rar1_GEL_R	Analytical replicate	”	”	”	”	27	27	0	50.0	
Rar2_GEL_R	Gizo 2	Coastal reference (mangrove)	*Geloina coaxans*	8º05'19"S	156º50'10"E	1.5	2	0	50.0	

Rar3_SAC_H	NGI (New Georgia Island)	Harbour	*Saccostrea sp*	8º14’08"S	157º11'45"E	0	1.5	12	81.9	50- >90
NZ1_MYT_R	Christchurch CE-1	Coastal reference	*Mytilus sp*	43º33'24"S	172º43'30"E	4	5	47	41.4	35-50
NZ2_MYT_M	Christchurch CE-3	Costal with ship traffic	*Mytilus sp*	43º33'51"S	172º45'0"E	2	3	30	57.5	55 - 80
NZ3_MYT_M	Christchurch CE-2	Costal with ship traffic	*Mytilus sp*	43º33'33"S	172º42'36"E	0.5	1.5	20	44.8	30-55
Chi1_MYT_R	Chile Skærgård	Costal refence	*Mytilus sp*	50º21'0"S	74º50'0"W	100	12	20	69.3	55- 90
Chi2_MYT_R	Isla Chiloe	Kyst reference	*Mytilus sp*	42º10'0"S	72º36'0"W	72	60	25	65.1	55-75
USV1_MYT_R	Salt-1, St. Croix	Coastal reference (mangrove)	*Brachidontes exustus*	17º46'16"N	64º45’09"W	1	4	309	28.8	20 - 40
USV1_ISO_R	Species replicat	”	*Isognomon alatus*	”	”	1	4	254	52.6	15-80
USV3_ISO_M	Hovensa, St. Croix	Estuarie with ship traffic	*Isognomon alatus*	17º41'44"N	64º45'50"W	2	1.5	186	35.4	20-70
USV2_ISO_H	Salt-3, St. Croix	Marina (Mangrove)	*Isognomon alatus*	17º46'31"N	64º45'40"W	0.1	0.1	57	41.0	20-60

Bos1_MYT_M	Boston-2	Outer pier	*Mytilus edulis*	2°22'14''N	71°03'29''W	0.1	0.1	63	51.2	35-65
Bos3_MYT_H	Boston-5	Old industrial Harbour	*Mytilus edulis*	42°21'47'N	71°01'50''V	0.5	5	44	56.7	40-70
Bos2_MYT_M	Boston-4	Coastal with ship traffic	*Mytilus edulis*	42°20'21	71°00'38''W	7.0	0.5	26	63.5	55-85
OW1_CHL_R	New Foundland Banq-1	Reference	*Chlamys islandica*	44º20'0''N	58º28'0''W	100	125	9	95.8	85->90
OW1_CHL_R	”	”	*Chlamys islandica*	”	”	100	125	9	95.8	85->90
OW2_CHL_R	Shetland Pent-1	Reference	*Tapes pullastra*	58º49'49''N	3º02'44''W	100	4	13	51.0	30-65

#: number; n.a.: not available.; n.r.: Not recorded

Replicate explanations: *Species replicate*: Two species sampled at the same station; *Replicate*: Two individual samplings, individual pools and homogenisation (“true” replicate); *Analytical: replicate*: Two digestions of one pool of mussels after homogenisation;

Table SI-3. Phylogenetic tree of species analysed, mainly based on the ITIS system (www.itis.gov)

Kingdom	Animalia							
Phylum	Mollusca							
Class	Bivalvia							
Subclass	Pteriomorphia						Heterodonta	
Order	Mytiloida			Pterioida	Ostreoida		Veneroida	
Family	Mytilidae			Isognomonidae	Pectinidae	Ostreidae	Veneridae	Corbiculidae
Genus	Mytilus	Perna	Brachidontes	Isognomon	Chlamys	Saccostrea	Tapes	Polymesoda
Species sampled	*M. edulis,* *M. gallopro-vincialis,* *M. sp*	*P. perna*	*B. exustus*	*I. alatus*	*C. opercularis,* *C. islandica*	*S. glomerata*[*#] *S. sp*	*T. pullastra*	*P. coaxans*[*§]

*: Species name not found in ITIS

#: Endemic to New Zealand/Australia (www.molusca.co.nz; Gould, 1850)

§: Corbicula Clams, indo-pacific regi (*polymesoda (Geloina) coaxans* (zipcodezoo.com; Gmelin, 1791))

sp: Species only determined to Genus level.

Tabel SI-4. Microsoft Excel 2003 Correl matrix on un-normalised data (29 parameters, 89 individual samples).

	TS	*Cd GFAA*	*Zn*	*Cd*	*Hg FIMS*	*Hg*	*Pb*	*Cu*	*Zn*	*P*	*Ni*	*Mg*	*Al*	*Ca*
TS	1.000													
Cd GFAA	0.055	1.000												
Zn	0.055	0.491	1.000											
Cd	-0.066	0.753	0.410	1.000										
Hg FIMS	0.163	-0.051	-0.004	-0.065	1.000									
Hg	0.034	-0.061	-0.041	-0.072	0.978	1.000								
Pb	0.201	0.334	0.147	0.212	0.027	0.008	1.000							
Cu	0.197	-0.054	0.044	-0.096	0.000	-0.026	-0.065	1.000						
Zn	-0.064	0.236	0.656	0.278	-0.082	-0.084	0.118	0.402	1.000					
P	0.052	0.034	0.113	0.065	0.152	0.075	0.119	-0.054	-0.072	1.000				
Ni	-0.022	-0.049	-0.081	-0.039	0.050	0.034	-0.131	0.225	0.029	0.127	1.000			
Mg	0.035	-0.041	0.291	-0.089	0.026	0.014	-0.164	0.098	0.331	-0.125	-0.053	1.000		
Al	0.136	-0.101	0.006	-0.082	-0.016	-0.058	0.007	0.257	0.103	-0.020	0.345	0.061	1.000	
Ca	0.381	-0.020	0.070	-0.062	0.002	-0.046	-0.078	0.153	0.084	-0.172	0.345	0.496	0.485	1.000
Mn	-0.073	-0.112	0.200	-0.116	0.047	0.057	-0.078	0.416	0.591	-0.089	0.078	0.630	0.267	0.300
K	-0.196	0.013	-0.017	0.000	0.034	0.011	-0.083	-0.190	-0.234	0.430	-0.165	0.118	-0.156	-0.236
V	-0.019	0.016	0.006	0.042	-0.066	-0.007	-0.009	-0.013	0.116	-0.147	0.436	0.258	0.203	0.372
Cr	0.322	0.006	0.164	-0.012	0.143	0.095	-0.095	0.326	0.193	0.078	0.335	0.383	0.604	0.742
Co	-0.094	-0.224	-0.170	-0.186	-0.046	-0.045	0.050	0.416	0.036	0.104	0.295	-0.177	0.588	0.049
Fe	0.065	-0.198	-0.026	-0.190	-0.011	-0.022	-0.094	0.011	-0.016	-0.168	0.149	0.218	0.153	0.238
Fe	0.146	-0.020	0.026	-0.011	0.150	0.086	-0.091	0.073	-0.096	0.291	0.896	0.044	0.353	0.421
As	0.131	-0.116	-0.088	-0.083	-0.103	-0.137	0.126	0.090	0.107	0.069	0.089	-0.021	0.036	0.049
Sr	0.064	-0.015	0.028	-0.046	-0.088	-0.098	-0.058	0.033	0.092	-0.108	0.350	-0.022	0.379	0.524
Mo	-0.347	-0.035	-0.133	0.125	-0.073	0.011	-0.229	0.198	0.094	-0.265	0.173	0.007	0.075	0.015
Th	0.162	-0.071	-0.057	0.004	0.022	-0.003	0.147	-0.004	-0.048	0.342	-0.026	-0.190	0.310	-0.006
U	-0.523	0.070	0.085	0.092	-0.230	-0.091	-0.175	-0.064	0.179	-0.202	0.223	-0.033	0.057	-0.047
TBT	0.231	0.125	0.162	0.031	0.027	-0.010	0.068	0.490	0.267	0.224	0.156	-0.130	0.014	-0.032
DBT	0.272	0.084	0.067	-0.021	-0.011	-0.047	0.043	0.730	0.284	0.047	0.185	-0.133	0.037	-0.039
MBT	0.157	0.075	0.138	-0.015	0.024	-0.004	0.024	0.477	0.227	0.122	0.211	-0.118	-0.016	-0.048

Tabel SI-4, Continued

	Mn	*K*	*V*	*Cr*	*Co*	*Fe*	*Fe*	*As*	*Sr*	*Mo*	*Th*	*U*	*TBT*	*DBT*	*MBT*
Mn	1.000														
K	-0.263	1.000													
V	0.255	-0.279	1.000												
Cr	0.354	-0.119	0.230	1.000											
Co	0.141	-0.051	0.007	0.241	1.000										
Fe	0.139	-0.114	0.142	0.158	0.164	1.000									
Fe	0.000	-0.068	0.338	0.449	0.219	0.208	1.000								
As	-0.016	0.116	0.079	0.007	0.262	-0.094	0.048	1.000							
Sr	0.039	-0.107	0.333	0.325	0.098	0.094	0.241	0.099	1.000						
Mo	0.156	0.029	0.160	-0.012	0.196	0.057	-0.084	-0.066	0.208	1.000					
Th	0.113	-0.256	0.175	0.123	0.430	0.030	0.066	-0.046	-0.149	-0.143	1.000				
U	0.085	-0.047	0.366	-0.095	0.106	0.103	-0.021	-0.090	0.365	0.535	-0.203	1.000			
TBT	0.084	-0.156	-0.095	0.064	0.173	0.007	0.103	0.007	-0.104	-0.031	0.161	-0.103	1.000		
DBT	0.106	-0.241	-0.114	0.057	0.244	-0.007	0.058	-0.003	-0.094	0.052	0.042	-0.120	0.879	1.000	
MBT	0.049	-0.152	-0.125	0.035	0.161	-0.003	0.126	-0.043	-0.061	0.052	-0.049	0.011	0.895	0.856	1.000

Numeric colour coding: Green >0.8; yellow between 0.6 and 0.8; red above 0.5 but below 0.6

Table SI-5 Supplemental bivalve species information

Family	Species	Living conditions	Reproductive cycle
Mytilidae	*Mytilus edulis*	Epilithic, filter feeding. Usually 5-10 cm, but up to 20 cm. Common along the north seaHigh intertidal to shallow subtidal (ca. 5 m), attached by fibrous byssus threads to rocks and other hard substrate.[12]	Spawning season in north sea usully April to October, in France also February – March. In Australia a minor in June and main spawning in August to January. [2]
	Mytilus Galloprovincialis	Epilithic. Up to 15 cm, slightly larger than M. edulis, and difficult to distinguish by shell shape alone. Invasive species in most of the world due to ballast water and cultivation, native to the Mediterranean coast and the Black and Adriatic Seas. Thrives in sediment-free fast flowing waters, with stands exposure to air for 7 days.[13]	Spawning occours when the water temperature is highest. Hybridization between species occours. Major spawning occurs in December-March and June-July. [3]
	Perna perna	Epilithic and mangrove roots, filter feeder. 9 cm in intertidal zones, 12 cm in sublittoral zone. Salinity tolerance 15-50 psu, temperature 15-30°C.[14]	Major spawning April-August. [3]
	Brachidontes exustus	Epilithic and mangrove roots, Filter feeder. 4.6 cm.[15]	Brachidontes exustus has two spawning periods in the spring, between March and April and, in the fall, between September and November
Isognomonidae	*Isognomon alatus*	mangrove roots, Filter feeder. 9.5 cm. The flat tree oyster is found only in the tropical regions of the Atlantic Ocean and Gulf of Mexico (Hall 1985). Isognomon alatus occurs from central Florida to Bermuda, the Bahamas, West Indies, Caribbean Central America, and as far south as	Isognomon alatus participates in mass spawning events peaking after the onset of high rains when the salinity of seawater is lower, i.e. September-October is the main spawning events occurs, depending on the hurricane season until January.[4]

		Brazil (Mikkelsen and Bieler 2008). It is generally found along the sub-tidal and intertidal surfaces of mangrove roots, in particular the red mangrove tree (Rhizophora mangle L.), at depths of 0.5 – 12 m.[18]	
Pectinidae	*Chlamys opercularis*	Epibenthic. The queen scallop is found between tidemarks, to depths of 100 m and on sand or gravel, often in high densities. 9 cm. Intertidal, but mostly 10 to 100 m depth. Temperature tolerance is up to 25 °C.[16]	Minor summer spawning in June-July and a more intensive autumn spawning in September-October [5]
	Chlamys islandica	Epibenthic. 11 cm 10-100 m depth. Temperature range: arctic to 15 °C.[17]	Spat settles in October-november. Spawning observed in july and april, probably march-april when ice breaks and phytoplankton blooms occur together with temperature variations from vernal meltwater discharges [6]
Ostreidae	*Saccostrea glomerata*	Epilithic, filter feeding. Up to 13 cm. Living intertidal to 3 m depth. Salinity range 0 to 38 psu (>20 psu for spat development), 10-30°C [7]	Peak spawning November to March, with some locations also in April, May.[8]
Veneridae	*Tapes pullastra*	Infaunal, Coastal species in sandy bottoms. from a few meters depth to ca 40 m, dual long siphons, detritus and phytoplankton feeder. Preferes saline waters. up to 6 cm.[19]	During summer. Usually separate sexes, with infrequent hermaphrodites. Spawning induced by temperature rise.
Corbiculidae	*Geloina coaxans*	Infaunal, prefers mudbanks. 15 cm.[20]	Spawning in India periode June – October, with peak in August – September. Maturing starts with temperature rise (October- January), and spawning occurs when salinity and temperature falls [9]

Table SI-6 Supplemental site information on hydrographic conditions and estimation of reproductive cycle status.

Site	**Hydrographic conditions[§] and date**	**Reproductive cycle**
1. Copenhagen	Most sampled in brackish to low salinity water in the Sound area and beneath. 10 -24 psu	All samples was taken outside of the spawning season.
2. Faroe Islands	17-18 August 2006: T 10-12°C, 35 psu	Outside Islandic *Mytilus edulis* spawning season. [10]
3. Greenland	22 August – 2 September: T 2-5°C, 10 psu (Grl4) T 2-5°C, 23 psu (Grl2) T 5-7°C, 24-27 psu (Grl 6, 7) 6-7°C, 32 psu (Grl1, 3, 5)	Outside Islandic *Mytilus edulis* and *Chlamys islandica* spawning season. [10]
4. Accra	2-10 October: T 26-27°C, 35-36 psu	Upper temperature limit for spawning of *Mytilus edulis*
5. Cape Town	15-18 October: T 11-14°C, 35 psu	Outside *Perna perna* spawning season
6. Perth	19-23 November: T 20-21°C, 36 psu Salinity in summer (October forward) is high (30 psu) at all stations, in winter salinity decreases as freshwater streams out. PER6 most affected, decreasing effect outward	All collected by hand during low tide, except the Coombe, collected by diver at 6 m depth Middle of spawning season
7. Hobart	6-8 December: 13-14°C, 34-35 psu	Last half of spawning season
8. Sydney	9-14 December: 21-23°C, 34-35 psu	Last half of spawning season
9. Gizo/Rarumana	28-31 December: 29-30°C, 34-35 psu	
10. Christchurch	7 January 2007: 14-15°C, 34 psu	End of spawning season
11. Chile	3 February: 17-18°C, 22 psu (Chi1) 17-18°C, 24-28 psu (Chi2)	After spawning season for Australia
12. St. Croix	21-29 March: 26-27°C, 36 psu	Probably in spawning season for *Brachidontes exustes*, Outside of spawning season of *Isognomon alatus*
13. Boston	11-14 April: 4-5°C, 30-33 psu	Outside of spawning season
14. Open water	16 April: 0-1°C, 32-35 psu (OW1) 23 April: 8-9°C, 34-35 psu (OW2)	Outside of spawning season for both chlamys species sampled

§: All dates, salinity and temperature data from Satelite Eye for Galathea [11]

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