Marine Pollution Bulletin
January 2017, Volume 114, Issue 2, Pages 1096-1102
http://dx.doi.org/10.1016/j.marpolbul.2016.10.018
http://archimer.ifremer.fr/doc/00353/46465/

Spatial and temporal distribution of mercury and methylmercury in bivalves from the French coastline

Briant Nicolas [1, *], Chouvelon Tiphaine [1], Martinez Laura [1], Brach-Papa Christophe [1], Chiffoleau Jean-Francois [1], Savoye Nicolas [2], Sonke Jeroen [3], Knoery Joel [1]

[1] Laboratoire de Biogéochimie des Contaminants Métalliques, Ifremer, Centre Atlantique, F-44311, Nantes Cedex 3, France
[2] Université Bordeaux, EPOC, UMR 5805, F-33400 Talence, France
[3] Observatoire Midi-Pyrénées, Laboratoire Géosciences Environnement Toulouse, CNRS/IRD/université Paul-Sabatier, 14, avenue Edouard-Belin, F-31400 Toulouse, France

* Corresponding author : Nicolas Briand

Abstract :

Marine mercury (Hg) concentrations have been monitored in the French coastline for the last half a century using bivalves. The analyses presented in this study concerned 192 samples of bivalves (mussels: Mytilus edulis and Mytilus galloprovincialis and oysters: Crassostrea gigas and Isognomon alatus) from 77 sampling stations along the French coast and in the French Antilles sea. The goals of this study were to assess MeHg levels in various common bivalves from French coastline, and to identify possible geographic, taxonomic or temporal variations of concentrations. We show that the evolution of methylmercury (MeHg) concentrations covary with total mercury (HgT) concentrations. Moreover, in most of the study sites, HgT concentrations have not decreased since 1987, despite regulations to decrease or ban mercury used for anthropic activities.

Highlights

► Hg concentrations did not decrease over the last 30 years. ► MeHg concentrations increase with total mercury concentrations. ► Time series of biological samples enable to quantify the evolution of pollutant levels in the coastal environment.

Keywords : Mercury, Methylmercury, France, Bivalves, Speciation

Mercury (Hg) is a non essential and highly toxic trace element for which the biogeochemical cycle is perturbed by anthropic inputs. Studies show that atmospheric depositions are the primary sources of mercury pollution in aquatic systems (Semkin, Mierle, et Neureuther 2005), and that anthropogenic activities continue to emit significant amounts of mercury into the air (Wu et al. 2006; Chen et al. 2013). Mercury levels have risen in the environment and have tripled in surface ocean waters since the industrial revolution (Lamborg et al. 2014). The toxicity of inorganic mercury causes renal lesions, neurotoxicity and cardiovascular disorders. Moreover the organic form of mercury (Methylmercury, MeHg) is the most toxic species for humans, causing serious central nervous system dysfunctions (Harada, 1995). Recently, the UNEP Governing Council drew up the Minamata Convention on Mercury (2013). This multilateral environmental agreement addresses the adverse effects of mercury through practical actions to protect human health and the environment from anthropogenic emissions and releases of mercury and mercury compounds. Monitoring by the French Mussel Watch program (RNO-ROCCH) can be useful to determine the effectiveness of these regulations. Indeed bivalves can accumulate pollutants in their tissues at elevated levels related to pollutant availability in the marine environment. This, they have been used as biomonitors of the coastal environment (« OSPAR Convention » 1992). As such, The French National Monitoring Network RNO-ROCCH (Observation Network of chemical contamination of the marine environment) aims to track chemical contaminants on the national coastline (Claisse 1989). Because not one single species of bivalve is common to all coastal regions, three species are collected to obtain national coverage.

The purposes of this study were to assess MeHg values in various common bivalves from French coastline and to identify possible geographic, taxonomic (intercomparison between species) or temporal variations of concentrations. We measured the MeHg concentrations of soft tissues in mussels (*Mytilus edulis, Mytilus galloprovincialis*) and oysters (*Crassostrea gigas, Isognomon alatus*). They were collected during the first trimester of years between 1987 and 2014 at various sites along the French shoreline and in French Caribbean islands. The objective was (i) to assess the spatial variation by determining MeHg concentrations of bivalves during one year at the same trimester around the French coasts (ii) to evaluate interspecific differences and linkage to local ecosystem characteristics, and (iii) to follow the evolution of MeHg concentrations over 30 years in sites of interest to check an eventual global tendency.

This study presents 192 samples of bivalves (mussels: *Mytilus edulis* and *Mytilus galloprovincialis* and oysters: *Crassostrea gigas* and *Isognomon alatus*) from 77 sampling stations along the French coast and in French Caribbean (Fig. 1). The spatial distribution study was realized on 75 sites of the metropolitan coast line with bivalves collected during the first semester (February and March) of 2014, to avoid seasonal variations of bivalves' tissues. The choice of the first semester was done by the ROCCH team because it is the period when the biological conditions of bivalves are the most stable. Two sites of French Caribbean islands were also sampled from the first semester of 2006 using local oyster species *Isognomon alatus*. Finally, the temporal variations were studied at 11. One "mussel" sample represents fifty individual mussels (35-65 mm in shell length) and one "oyster" sample represents 10 cultivated bivalves between two and three years old. Bivalves have been collected in an identical *modus operandi* every year since 1979 within the framework of the French mussel watch program (RNO-ROCCH). Briefly, they are cleaned of epibiota and depurated for 24 h in a PE tank containing decanted water from the sampling site, in order to eliminate faeces and pseudofaeces. The soft tissue is removed from the shell, homogenized with a stainless steel blade, and freeze-dried as recommended by the guidelines of the OSPAR Convention. The method is fully described in Claisse (1989). The MeHg determination was performed according to Azemard et Vassileva (2015) using a liquid-liquid extractions. Analyses were carried out using an advanced mercury analyzer (AMA-254, Altech Czech Republic). After solubilization of the stored lyophilized tissues in HCl (3N), toluene is added and both phases homogenized. After centrifugation, a fraction of the upper organic phase with extracted MeHg is transferred to a second tube containing the same volume of 2mM sodium thiosulfate solution. This second tube was vigorously shaken and centrifuged. An aliquot (400µL) of the lower phase, which contains the back-extracted organic mercury, was directly analyzed with the AMA. Precision was determined by comparing the analyzed concentrations of total Hg (HgT) (61.00 ± 3.60 $\mu g.kg^{-1}$ d.w) and MeHg (28.09 ± 0.31 $\mu g.kg^{-1}$ d.w) with the certified values of the CRM-NIST-2976. For MeHg, our CRM mean value (n=36) is 27.83 ± 0.22 $\mu g.kg^{-1}$ d.w. The detection limit of the AMA-254 was 0.05 ng of Hg. HgT was directly analyzed by the AMA from the lyophilized samples. Statistical analyses were done using the free software "R". Because the data were not normally distributed, nonparametrical statistical approaches were utilized for data comparison.

Concentrations of HgT and MeHg in oyster and mussel tissues are presented in Table I. Concentrations of HgT and MeHg of the French coast line in 2014 varied between 60 and 670µg.kg^{-1} d.w and 9 to 145µg.kg^{-1} d.w respectively. MeHg values were within the range of values reported previously in other studies (Odžak et al. 2000; Claisse et al. 2001; Apeti, Lauenstein, et Evans 2012). Comparisons of values between species are done regarding results from statistical tests. For HgT and for MeHg level the sole interspecific differences were observed between *C.gigas* and *Mytilus spp*. *Isognomon alatus* values are too few to be statistically used. After combining all sampling sites, mean concentration of HgT are similar for both mussels species (Wilcoxon test, *p*value = 0.31). Mean concentration is different for oysters *C.gigas* (Wilcoxon test, *p*value <0.05) with both mussels species. HgT concentrations for *C.gigas* are slightly higher than those of *Mytilus spp* (240µg.kg-1 d.w versus ~174 µg.kg-1 d.w respectively). Same tests for MeHg concentrations show no difference between species (Wilcoxon test, *p*value > 0.05). Levels of HgT and MeHg in bivalves revealed a wide variability in the distribution of mercury across the monitoring area. Statistical analyses of HgT or MeHg levels did not permit to differentiate specific ecosystem specificities such as estuarine or bay or open coast or lagoon.

Nevertheless different “hotspots” could be observed for HgT and MeHg. We defined hotspots as top 10% of the concentration distribution. They have levels above 320µg.kg-1 d.w for HgT and above 103µg.kg-1 d.w for MeHg. There is evidence of the influence of mercury point sources near several estuaries. Elevated concentrations of HgT are found in bivalves of Seine Bay, near the Loire estuary and in the Gironde estuary (320, 360, 380 µg.kg-1 d.w respectively, Fig.2). For some estuaries such as the Seine historical industrial activity could explain the quite high concentration of Hg (Foucher 2002 ; Laurier et al. 2003). As described by Claisse et al. (2001), the main contamination spot is Toulon-Lazaret site (#71, Fig.1) with concentrations up to 670µg.kg-1 d.w of HgT and 132µg.kg-1 d.w of MeHg. This bay is well known as a highly contaminated site, most probably linked to military activities of this harbor (Tessier et al. 2011). Moreover, Tessier et al. (2011) have identified mercury as the most problematic contaminant of this site, with wide dispersion throughout the whole bay.

Except for Toulon-Lazaret which is highly contaminated, MeHg “hotspots” are quite unusual from our knowledge. In the English Channel coast at Le Moulard site (#13), MeHg concentration is 135µg.kg-1 d.w, which represents 48% of HgT. The other hotspot is

Noirmoutier site (#39) at the south of the Loire River estuary. The levels are ~145µg.kg-1 d.w which represent 40% of the HgT concentration. For these two sites, we have not observed an obvious source of this contamination, although local over-productivity of MeHg by biotic or abiotic processes in nearly salt marshes might be an explanation for these results (Mitchell and Gilmour. 2008).

Methylmercury levels in bivalve soft tissues varied greatly along the French coast, yet no significant differences were found between organisms (Table I). The proportion of MeHg to HgT ranged from 13.1 to 54.1%, with a median of 30.4%. The highest percentages corresponded to the sites Varengeville (#5) and Le Moulard (#13) in the English Channel coast, Brittany sites Pointe er Fosse (#34) and Le Croisic (#36) for the Atlantic coast, and the Prévost lagoon (#65) on the Mediterranean coast. Ratios ranged over 48% and up to 54.1% at Varengeville. This latter particular site was already described with high mercury and methylmercury concentrations coming from the karstic watershed of the Caux region (Laurier et al. 2007). Inputs from groundwater with high MeHg concentrations were also shown by Ganguli et al. 2012 in a coastal lagoon system. In this study, the MeHg/HgT ratios are lower than previous studies on the French coast. Claisse et al. (2001) reported ratio of about 43%, Mikac et al. (1996) reported ratio about 40% in mussels from the Kska Estuary in Croatia. However Apeti et al, 2012 reported a median ratio of 34% in oysters of the northern Gulf of Mexico. Generally, MeHg concentrations increase with HgT in the environment (Fig 2c). How does MeHg levels depend on Hg concentrations?

Total mercury concentrations are still high in many point of the French coast despite the decrease of Hg usage for various anthropic activities (phase out of chlor-alkali plants, mercury batteries or medical thermometers, etc). Indeed the mercury concentrations in a wide variety of ecosystems and geographic localization do not show a significant decrease over the last 30 years (Fig. 3).

For the three studied organisms, HgT concentrations as well as MeHg concentrations did not decrease since 1987. Moreover in the case of Villerville (#9) and Toulon–Lazaret a slight increase could be observed between 1987 and 2014. As explained before, Toulon Lazaret is a highly contaminated site and the increase of concentrations could be explained by local remobilization of dredged sediment from the inner bay. In the case of Villerville, the Seine River could be bringing to that site more Hg year after year. Although, the concentration patterns of MeHg and HgT look similar, the fraction of MeHg fluctuates independently between 20 and 50%. These observations suggest that HgT is not the limiting factor in the environment for the production of MeHg, and that other factors like bacterial methylation activity, organic matter quality and quantities or sulfate concentrations are more limiting (Choi et Bartha 1994; Heyes, Miller, et Mason 2004). However in a context of global change and an increase of ocean temperatures, one or more of these factors may become less limiting, and potentially raising future MeHg concentrations at the base of the food web.

This study showed that long time series of biological samples are crucial to follow the evolution of pollutant levels. Moreover we identified that in the majority of the study sites HgT concentrations have not decreased significantly since 1987, despite national, European and international regulation to decrease mercury used in anthropic activities, and despite the decrease in Hg atmospheric deposition since the 1990's (Zhang et al. 2016). Since oceanic and coastal ecosystems are complex with respect to Hg behavior and fate, a lag could occur between atmospheric deposition and ocean concentrations (Sunderland et Mason 2007). Bivalves represent the base of the trophic food web, and MeHg concentrations ranged from 9 to 145$\mu g.kg^{-1}$. With the risk of global change -mediated changes of MeHg levels, the origin of mercury introduced into coastal food web needs to be determined, and parameters like $\delta^{15}N$, $\delta^{13}C$ and Hg stable isotopes could be useful.

Acknowledgement

We gratefully acknowledge funding of this study obtained from IFREMER, Région Pays de la Loire (PolluSols), and the INSU program EC2CO/DRIL.

Captions

Table I: Sample table for the 2014 mercury speciation map. Concentrations and summary of site characteristics.

Fig 1: Geographic location of the sampling sites.

Fig 2: a) Spatial distribution of the mercury in the bivalves. Blue dots are the exact locations and symbol shapes and colors identify the type of organisms. **b)** Spatial distribution of methylmercury in bivalves. **c)** Correlations of methylmercury (MeHg) with total mercury (HgT) for *C.gigas* and *Mytilus spp* for metropolitan France in 2014. Geographic location of the sampling sites.

Fig 3: Concentrations of HgT (open diamonds) and MeHg (black dots) at eleven sites from metropolitan France, from 1987 to 2014 with a step 3 years time. Also shown are the fitted curves from a logistic Generalised Linear Model.

References:

Apeti, D. A., G. G. Lauenstein, et D. W. Evans. 2012. « Recent status of total mercury and methyl mercury in the coastal waters of the northern Gulf of Mexico using oysters and sediments from NOAA's mussel watch program ». *Marine pollution bulletin* 64 (11): 2399‑ 2408.

Azemard, S, et E Vassileva. 2015. « Determination of methylmercury in marine biota samples with advanced mercury analyzer: Method validation ». *Food chemistry* 176: 367‑ 75.

Chen, L, M Liu, R Fan, S Ma, Z Xu, M Ren, et Q He. 2013. « Mercury speciation and emission from municipal solid waste incinerators in the Pearl River Delta, South China ». *Science of The Total Environment* 447 (mars): 396‑ 402.

Choi, S.-C., et R. Bartha. 1994. « Environmental factors affecting mercury methylation in estuarine sediments ». *Bulletin of Environmental Contamination and Toxicology* 53 (6): 805‑ 12.

Claisse, D. 1989. « Chemical contamination of French coasts: the results of a ten years mussel watch ». *Marine pollution bulletin* 20 (10): 523‑ 28.

Claisse, D., D. Cossa, J. Bretaudeau-Sanjuan, G. Touchard, et B. Bombled. 2001. « Methylmercury in molluscs along the French coast ». *Marine pollution bulletin* 42 (4): 329‑ 32.

Foucher, D. 2002. « Geochemistry of mercury in estuarine and coastal sediments: case studies in the Seine estuary (France) and the Kastela bay (Croatia) ». Ph. D. thesis. Univ. Lille 1, France.

Ganguli P. M., C. H. Conaway, P. W. Swarzenski, J. A. Izbicki and A. R. Flegal. 2012 « Mercury Speciation and Transport via Submarine Groundwater Discharge at a Southern California Coastal Lagoon System ». *Environmental Science & Technology*, 46, 1480‑ 88

Harada, M. 1995. « Minamata Disease: Methylmercury Poisoning in Japan Caused by Environmental Pollution ». *Critical Reviews in Toxicology* 25 (1): 1‑ 24.

Heyes, A, C Miller, et R P Mason. 2004. « Mercury and methylmercury in Hudson River sediment: impact of tidal resuspension on partitioning and methylation ». *Marine Chemistry* 90 (1): 75‑ 89.

Lamborg, C H., C R. Hammerschmidt, K L. Bowman, G J. Swarr, K M. Munson, D C. Ohnemus, P J. Lam, L-E Heimbürger, M J. A. Rijkenberg, et M A. Saito. 2014. « A Global Ocean Inventory of Anthropogenic Mercury Based on Water Column Measurements ». *Nature* 512 (7512): 65‑ 68. doi:10.1038/nature13563.

Laurier, F. J. G., D Cossa, J. L Gonzalez, E Breviere et G Sarazin. 2003. « Mercury transformations and exchanges in a high turbidity estuary: The role of organic matter and amorphous oxyhydroxides ». *Geochimica et Cosmochimica Acta* 67 (18): 3329‑ 45

Laurier, F. J. G., D Cossa, C Beucher, et E Breviere. 2007. « Reprint of "The impact of groundwater discharges on mercury partitioning, speciation and bioavailability to mussels in a coastal zone" ». *Marine Chemistry* 106 (1): 352‑ 64.

Mikac, N, Ž Kwokal, D Martinčić, et M Branica. 1996. « Uptake of mercury species by transplanted mussels Mytilus galloprovincialis under estuarine conditions (Krka river estuary) ». *Science of the total environment* 184 (3): 173‑ 82.

Mitchell, C. P. J., et C. C. Gilmour 2008. Methylmercury production in a Chesapeake Bay salt marsh, *Journal of Geophysical Research*, 113

Odžak, N, T Zvonarić, Z K Gašpić, M Horvat, et A Barić. 2000. « Biomonitoring of mercury in the Kaštela Bay using transplanted mussels ». *Science of the total environment* 261 (1): 61‑ 68.

OSPAR Convention-1992. *OSPAR Commission*. http://www.ospar.org/convention/text.

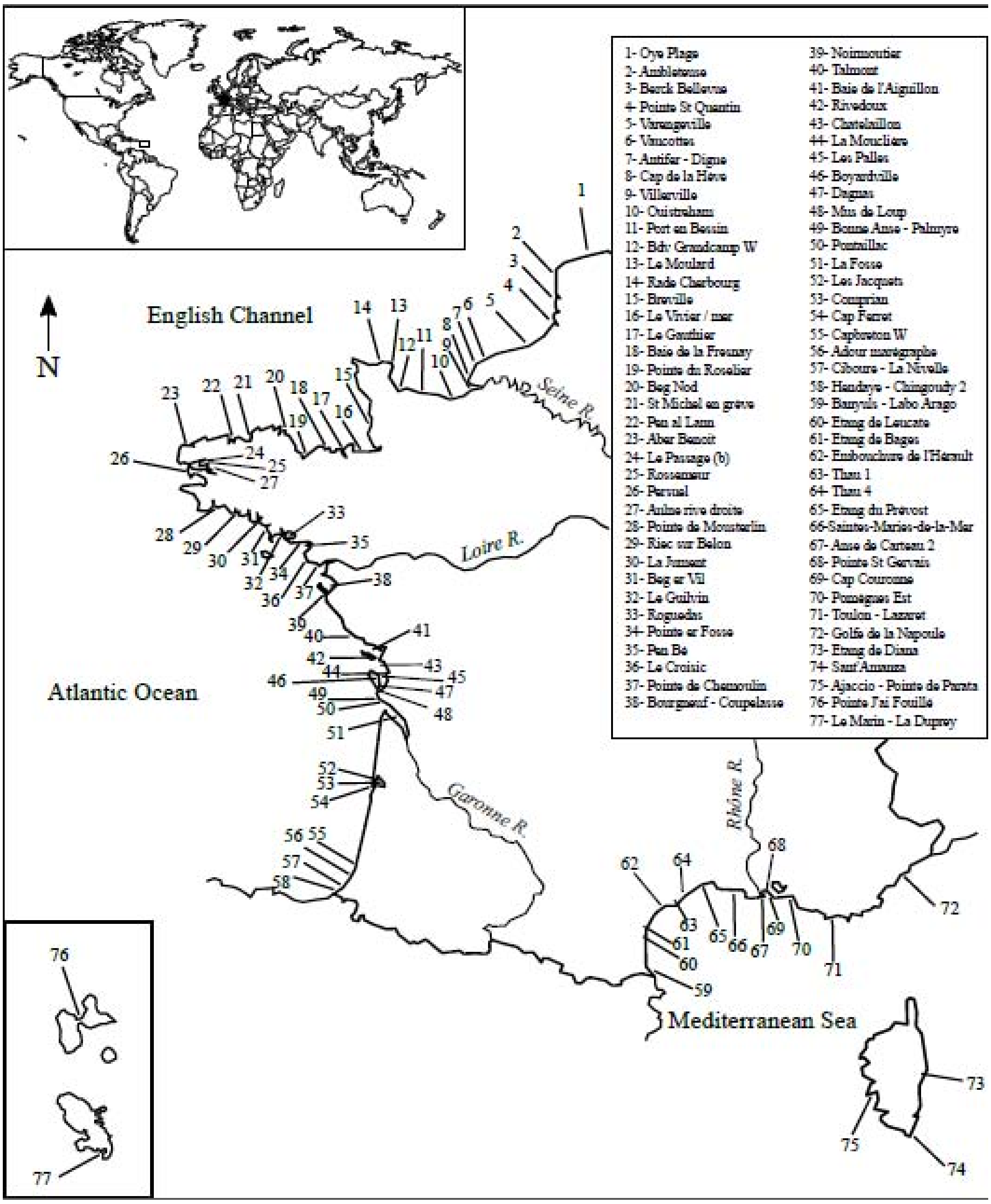
Semkin, R G., G Mierle, et R J. Neureuther. 2005. « Hydrochemistry and Mercury Cycling in a High Arctic Watershed ». *The Science of the Total Environment* 342 (1-3): 199‑ 221.

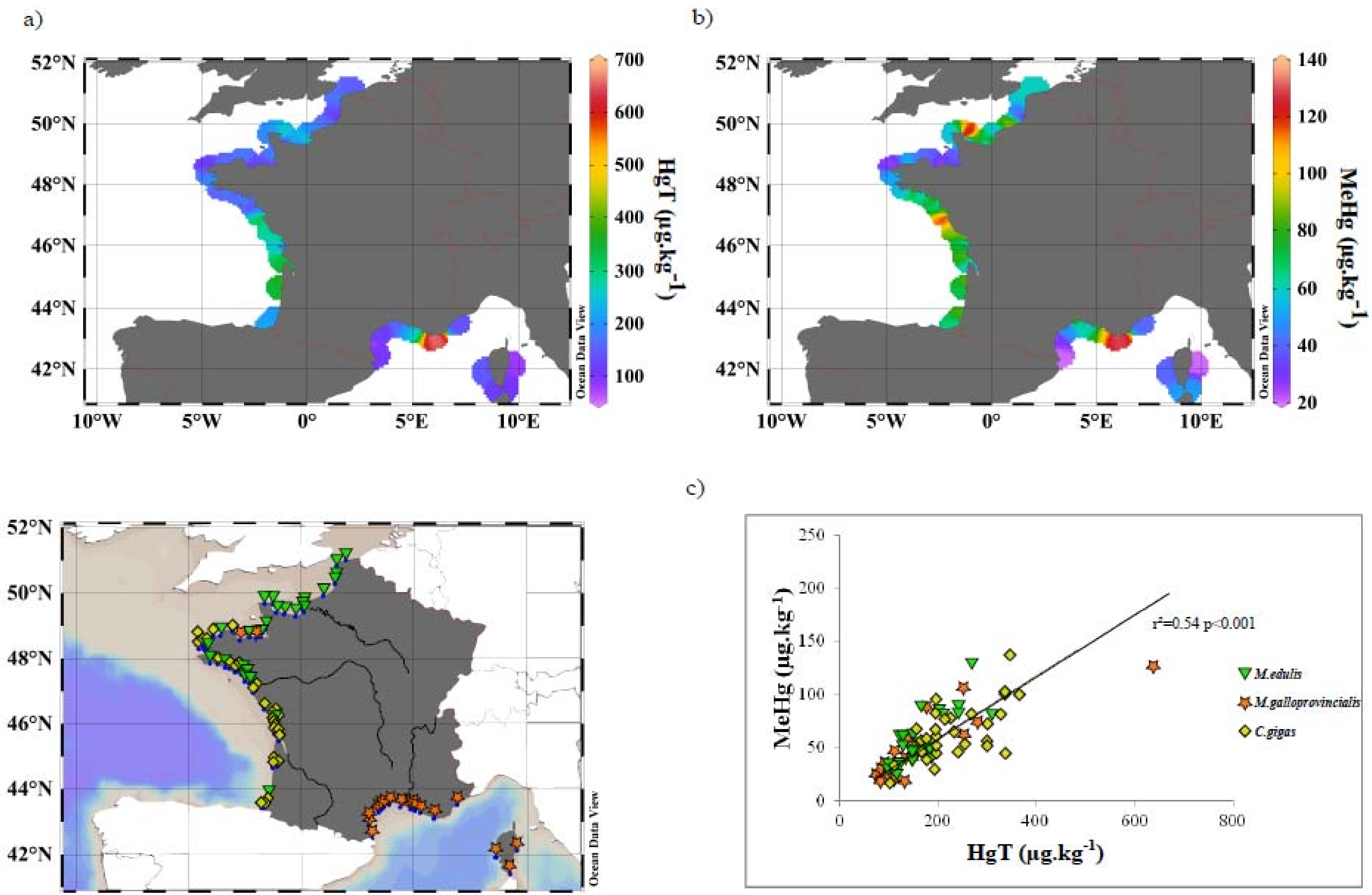
Sunderland, E. M., et R. P. Mason. 2007. « Human impacts on open ocean mercury concentrations ». *Global Biogeochemical Cycles* 21 (4).

Tessier, E, C Garnier, J-U Mullot, V Lenoble, M Arnaud, M Raynaud, et S Mounier. 2011. « Study of the spatial and historical distribution of sediment inorganic contamination in the Toulon bay (France) ». *Marine pollution bulletin* 62 (10): 2075‑ 86.

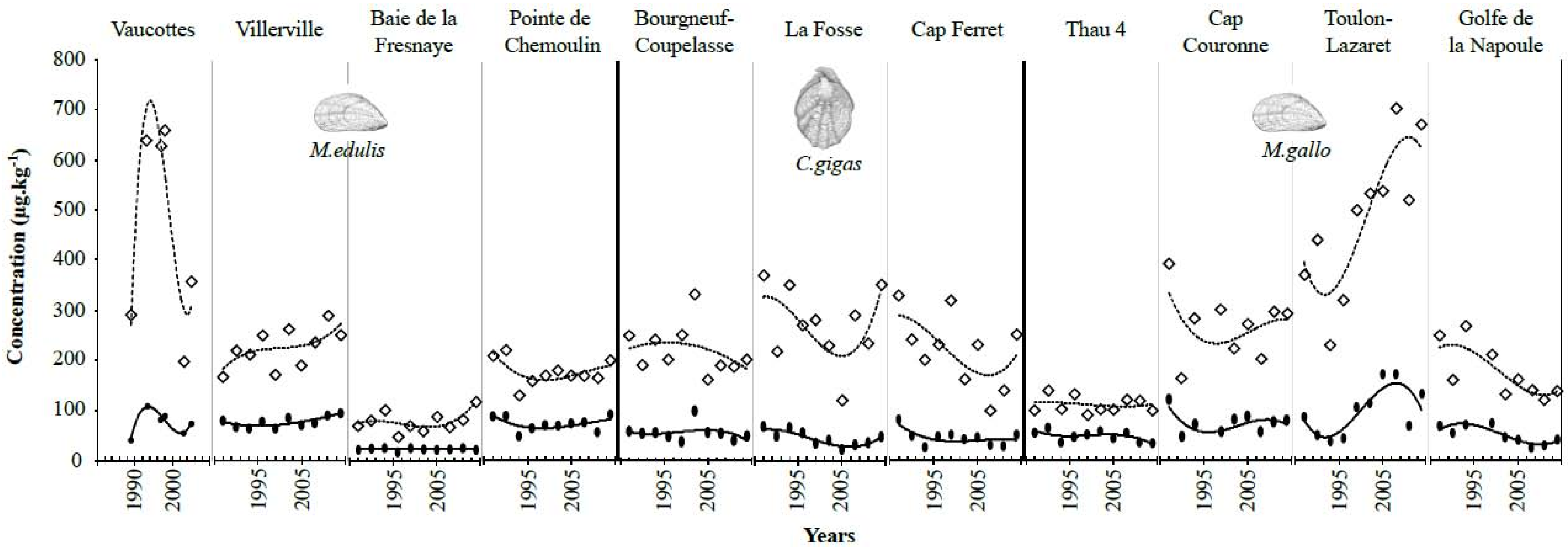
Wu, Y, S Wang, D G Streets, J Hao, M Chan, et J Jiang. 2006. « Trends in Anthropogenic Mercury Emissions in China from 1995 to 2003 ». *Environmental Science & Technology* 40 (17): 5312‑ 18.

Zhang, Y, D. J. Jacob, H. M. Horowitz, L. Chen, H. M, Amos, D. P. Krabbenhoft, F. Slemr, V. L. St Louis et E. M. Sunderland. 2016. « Observed decrease in atmospheric mercury explained by global decline in anthropogenic emissions ». *Proceeding of the National Academy of Science of United States of America* 113 (3): 526‑31.


English Channel
N
Atlantic Ocean
Mediterranean Sea
Seine R.
Loire R.
Garonne R.
Rhône R.
1- Oye Plage
2- Ambleteuse
3- Berck Bellevue
4- Pointe St Quentin
5- Varengeville
6- Vaucottes
7- Antifer - Digue
8- Cap de la Hève
9- Villerville
10- Ouistreham
11- Port en Bessin
12- Bdv Grandcamp W
13- Le Moulard
14- Rade Cherbourg
15- Breville
16- Le Vivier / mer
17- Le Gauthier
18- Baie de la Fresnay
19- Pointe du Roselier
20- Beg Nod
21- St Michel en grève
22- Pen al Lann
23- Aber Benoit
24- Le Passage (b)
25- Rossemeur
26- Persuel
27- Aulne rive droite
28- Pointe de Mousterlin
29- Riec sur Belon
30- La Jument
31- Beg er Vil
32- Le Guilvin
33- Roguedas
34- Pointe er Fosse
35- Pen Bé
36- Le Croisic
37- Pointe de Chemoulin
38- Bourgneuf - Coupelasse
39- Noirmoutier
40- Talmont
41- Baie de l'Aiguillon
42- Rivedoux
43- Chatelaillon
44- La Moulière
45- Les Palles
46- Boyardville
47- Dagnas
48- Mus de Loup
49- Bonne Anse - Palmyre
50- Pontaillac
51- La Fosse
52- Les Jacquets
53- Comprian
54- Cap Ferret
55- Capbreton W
56- Adour marégraphe
57- Ciboure - La Nivelle
58- Hendaye - Chingoudy 2
59- Banyuls - Labo Arago
60- Etang de Leucate
61- Etang de Bages
62- Embouchure de l'Hérault
63- Thau 1
64- Thau 4
65- Etang du Prévost
66-Saintes-Maries-de-la-Mer
67- Anse de Carteau 2
68- Pointe St Gervais
69- Cap Couronne
70- Pomègues Est
71- Toulon - Lazaret
72- Golfe de la Napoule
73- Etang de Diana
74- Sant'Amanza
75- Ajaccio - Pointe de Parata
76- Pointe J'ai Fouillé
77- Le Marin - La Duprey



a)
b)
c)
52°N
50°N
48°N
46°N
44°N
42°N
10°W
5°W
0°
5°E
10°E
HgT (µg.kg-1)
MeHg (µg.kg-1)
Ocean Data View
r²=0.54 p<0.001
M.edulis
M.galloprovincialis
C.gigas



Vaucottes
Villerville
Baie de la Fresnaye
Pointe de Chemoulin
Bourgneuf-Coupelasse
La Fosse
Cap Ferret
Thau 4
Cap Couronne
Toulon-Lazaret
Golfe de la Napoule
M.edulis
C.gigas
M.gallo
Concentration (µg.kg-1)
800
700
600
500
400
300
200
100
0
1990
2000
1995
2005
Years

Site	Specie	Sea/Ocean location	Ecosystem	Year	HgT (µg.kg^{-1} dw)	MeHg (µg.kg^{-1} dw)	IHg (µg.kg^{-1} dw)	ratio (%)
Oye plage	*M. edulis*	English Channel	Coast	2014	150	64	86	42.5
Ambleteuse	*M. edulis*	English Channel	Coast	2014	180	56	124	30.9
Berck Bellevue	*M. edulis*	English Channel	Coast	2014	150	49	101	32.5
Pointe de St Quentin	*M. edulis*	English Channel	Coast	2014	100	26	74	26.3
Varengeville	*M. edulis*	English Channel	Coast	2014	170	92	78	54.1
Antifer - digue	*M. edulis*	English Channel	Coast	2014	170	52	118	30.7
Cap de la Hève	*M. edulis*	English Channel	Estuary	2014	320	85	235	26.5
Ouistreham	*M. edulis*	English Channel	River	2014	150	48	102	31.9
Villerville	*M. edulis*	English Channel	Estuary	2014	250	94	156	37.6
Port en Bessin	*M. edulis*	English Channel	Coast	2014	250	85	165	34.2
Bdv Grandcamp ouest	*M. edulis*	English Channel	River	2014	100	30	70	30.0
Le Moulard	*M. edulis*	English Channel	Coast	2014	280	135	145	48.2
Grande rade de Cherbourg	*M. edulis*	English Channel	Bay	2014	190	48	142	25.3
Bréville	*M. edulis*	English Channel	Coast	2014	190	40	150	21.1
Le Vivier sur mer	*M. edulis*	English Channel	Bay	2014	120	31	89	25.8
La Gauthier	*M. galloprovincialis*	English Channel	River	2014	160	46	114	28.8
Baie de la Fresnaye	*M. edulis*	English Channel	Bay	2014	120	22	98	18.3
Pointe du Roselier	*M. galloprovincialis*	English Channel	Coast	2014	90	26	64	28.9
Beg Nod	*C. gigas*	English Channel	Coast	2014	180	43	137	23.9
St Michel en grève	*M. edulis*	English Channel	Coast	2014	150	39	111	25.8
Pen al Lann	*C. gigas*	English Channel	Coast	2014	170	60	110	35.5
Aber Benoît	*C. gigas*	English Channel	Coast	2014	100	18	82	18.2
Le Passage (b)	*C. gigas*	Atlantic	Bay	2014	230	83	147	36.1
Rossermeur	*C. gigas*	Atlantic	Bay	2014	110	33	77	30.1
Persuel	*C. gigas*	Atlantic	Bay	2014	170	50	120	29.3
Aulne rive droite	*C. gigas*	Atlantic	River	2014	310	59	251	18.9
Pointe de Mousterlin	*M. edulis*	Atlantic	Coast	2014	150	49	101	32.4
Riec sur Belon	*C. gigas*	Atlantic	River	2014	200	89	111	44.3
La Jument	*M. edulis*	Atlantic	River	2014	130	61	69	46.6
Beg er Vil	*C. gigas*	Atlantic	River	2014	180	62	118	34.6
Le Guilvin	*C. gigas*	Atlantic	Bay	2014	200	72	128	35.8
Roguedas	*C. gigas*	Atlantic	River	2014	160	69	91	43.4
Pointe er Fosse	*C. gigas*	Atlantic	River	2014	200	100	100	49.9
Pen Bé	*M. edulis*	Atlantic	Bay	2014	130	53	77	40.8
Le Croisic	*M. edulis*	Atlantic	Coast	2014	120	61	59	51.0
Pointe de Chemoulin	*M. edulis*	Atlantic	Estuary	2014	200	92	108	46.1
Bourgneuf - Coupelasse	*C. gigas*	Atlantic	Bay	2014	200	48	152	24.1
Noirmoutier - Gresse-loup	*C. gigas*	Atlantic	Bay	2014	360	145	215	40.2
Talmont	*C. gigas*	Atlantic	River	2014	280	87	193	30.9
Rivedoux	*C. gigas*	Atlantic	Coast	2014	260	57	203	22.1
Baie de l'Aiguillon	*C. gigas*	Atlantic	Bay	2014	350	108	242	30.8
Châtelaillon	*C. gigas*	Atlantic	Coast	2014	200	32	168	16.0
Les Palles	*C. gigas*	Atlantic	River	2014	220	82	138	37.1
Boyardville	*C. gigas*	Atlantic	Coast	2014	350	111	239	31.8
Dagnas	*C. gigas*	Atlantic	Bay	2014	240	69	171	28.8
La Mouclière	*M. edulis*	Atlantic	River	2014	120	29	91	23.9
Mus de loup	*C. gigas*	Atlantic	River	2014	200	65	135	32.6
Pontaillac	*C. gigas*	Atlantic	Estuary	2014	310	55	255	17.8
Bonne Anse - Palmyre	*C. gigas*	Atlantic	Estuary	2014	380	106	274	27.9
La Fosse	*C. gigas*	Atlantic	Estuary	2014	350	48	302	13.6
Cap Ferret	*C. gigas*	Atlantic	Bay	2014	250	51	199	20.5
Les Jacquets	*C. gigas*	Atlantic	Bay	2014	340	87	253	25.6
Comprian	*C. gigas*	Atlantic	Bay	2014	310	77	233	24.9
Capbreton ouest	*M. edulis*	Atlantic	River	2014	210	89	121	42.3
Adour marégraphe	*C. gigas*	Atlantic	River	2014	200	56	144	28.1
Ciboure - la Nivelle	*C. gigas*	Atlantic	River	2014	260	57	203	21.9
Hendaye - Chingoudy 2	*C. gigas*	Atlantic	River	2014	200	70	130	34.8
Banyuls - Labo Arago	*M. galloprovincialis*	Mediterranean	Coast	2014	110	20	90	18.1
Embouchure de l'Hérault	*M. galloprovincialis*	Mediterranean	River	2014	140	61	79	43.4
Etang de Leucate	*M. galloprovincialis*	Mediterranean	Lagoon	2014	70	25	45	35.8
Etang de Bages	*M. galloprovincialis*	Mediterranean	Lagoon	2014	132	19	113	14.2
Thau 1	*M. galloprovincialis*	Mediterranean	Lagoon	2014	90	36	54	40.4
Thau 4	*M. galloprovincialis*	Mediterranean	Lagoon	2014	100	33	67	32.8
Etang du Prévost	*M. galloprovincialis*	Mediterranean	Lagoon	2014	180	91	89	50.7
Saintes-Maries-de-la-Mer	*M. galloprovincialis*	Mediterranean	Coast	2014	160	57	103	35.4
Pointe St Gervais	*M. galloprovincialis*	Mediterranean	Bay	2014	260	65	195	24.9
Anse de Carteau 2	*M. galloprovincialis*	Mediterranean	Bay	2014	170	51	119	29.9
Cap Couronne	*M. galloprovincialis*	Mediterranean	Coast	2014	290	77	213	26.7
Pomègues Est	*M. galloprovincialis*	Mediterranean	Coast	2014	260	112	148	43.2
Toulon - Lazaret	*M. galloprovincialis*	Mediterranean	Bay	2014	670	132	538	19.7
Golfe de la Napoule	*M. galloprovincialis*	Mediterranean	Coast	2014	140	41	99	29.3
Etang de Diana	*M. galloprovincialis*	Mediterranean	Lagoon	2014	80	18	62	22.9
Sant'Amanza	*M. galloprovincialis*	Mediterranean	Bay	2014	110	49	61	44.1
Ajaccio - Pte de Parata	*M. galloprovincialis*	Mediterranean	Coast	2014	150	41	109	27.6
Pointe J'ai Fouillé	*I.alatus*	Caribbean	Coast	2006	70	9	61	13.1
Le Marin - La Duprey	*I.alatus*	Caribbean	Coast	2006	60	17	43	27.5

Site	Specie	Sea/Ocean location	Ecosystem	Year	HgT ($\mu g.kg^{-1}$ dw)	MeHg ($\mu g.kg^{-1}$ dw)	IHg ($\mu g.kg^{-1}$ dw)	ratio (%)
Vaucottes	*M. edulis*	English Channel	Coast	1989	290	46	244	15.9
Vaucottes	*M. edulis*	English Channel	Coast	1993	640	109	531	17.1
Vaucottes	*M. edulis*	English Channel	Coast	1997	630	82	548	12.9
Vaucottes	*M. edulis*	English Channel	Coast	1998	660	90	570	13.6
Vaucottes	*M. edulis*	English Channel	Coast	2003	200	56	144	28.0
Vaucottes	*M. edulis*	English Channel	Coast	2005	360	77	283	21.3
Villerville	*M. edulis*	English Channel	Coast	1987	170	79	91	46.6
Villerville	*M. edulis*	English Channel	Coast	1990	220	66	154	30.1
Villerville	*M. edulis*	English Channel	Coast	1993	210	63	147	30.2
Villerville	*M. edulis*	English Channel	Coast	1996	250	77	173	30.9
Villerville	*M. edulis*	English Channel	Coast	1999	170	63	107	37.3
Villerville	*M. edulis*	English Channel	Coast	2002	260	85	175	32.6
Villerville	*M. edulis*	English Channel	Coast	2005	190	71	119	37.2
Villerville	*M. edulis*	English Channel	Coast	2008	240	74	166	31.0
Villerville	*M. edulis*	English Channel	Coast	2011	288	90	198	31.2
Villerville	*M. edulis*	English Channel	Coast	2014	250	94	156	37.6
Baie de la Fresnaye	*M. edulis*	English Channel	Bay	1987	70	22	48	31.7
Baie de la Fresnaye	*M. edulis*	English Channel	Bay	1990	80	24	56	30.6
Baie de la Fresnaye	*M. edulis*	English Channel	Bay	1993	100	26	74	25.5
Baie de la Fresnaye	*M. edulis*	English Channel	Bay	1996	50	17	33	34.2
Baie de la Fresnaye	*M. edulis*	English Channel	Bay	1999	70	25	45	36.3
Baie de la Fresnaye	*M. edulis*	English Channel	Bay	2002	60	23	37	38.4
Baie de la Fresnaye	*M. edulis*	English Channel	Bay	2005	90	22	68	24.6
Baie de la Fresnaye	*M. edulis*	English Channel	Bay	2008	70	23	47	32.9
Baie de la Fresnaye	*M. edulis*	English Channel	Bay	2011	83	25	58	30.6
Baie de la Fresnaye	*M. edulis*	English Channel	Bay	2014	120	22	98	18.3
Pointe de Chemoulin	*M. edulis*	Atlantic	Estuary	1987	210	88	122	42.0
Pointe de Chemoulin	*M. edulis*	Atlantic	Estuary	1990	220	89	131	40.5
Pointe de Chemoulin	*M. edulis*	Atlantic	Estuary	1993	130	49	81	37.3
Pointe de Chemoulin	*M. edulis*	Atlantic	Estuary	1996	160	64	96	40.2
Pointe de Chemoulin	*M. edulis*	Atlantic	Estuary	1999	170	71	99	41.7
Pointe de Chemoulin	*M. edulis*	Atlantic	Estuary	2002	180	70	110	38.8
Pointe de Chemoulin	*M. edulis*	Atlantic	Estuary	2005	170	74	96	43.8
Pointe de Chemoulin	*M. edulis*	Atlantic	Estuary	2008	170	76	94	44.9
Pointe de Chemoulin	*M. edulis*	Atlantic	Estuary	2011	166	57	109	34.2
Pointe de Chemoulin	*M. edulis*	Atlantic	Estuary	2014	200	92	108	46.1
Bourgneuf - Coupelasse	*C. gigas*	Atlantic	Bay	1987	250	57	193	22.9
Bourgneuf - Coupelasse	*C. gigas*	Atlantic	Bay	1990	190	53	137	28.1
Bourgneuf - Coupelasse	*C. gigas*	Atlantic	Bay	1993	240	55	185	22.8
Bourgneuf - Coupelasse	*C. gigas*	Atlantic	Bay	1996	200	47	153	23.5
Bourgneuf - Coupelasse	*C. gigas*	Atlantic	Bay	1999	250	37	213	14.6
Bourgneuf - Coupelasse	*C. gigas*	Atlantic	Bay	2002	330	98	232	29.6
Bourgneuf - Coupelasse	*C. gigas*	Atlantic	Bay	2005	160	55	105	34.4
Bourgneuf - Coupelasse	*C. gigas*	Atlantic	Bay	2008	190	53	137	28.1
Bourgneuf - Coupelasse	*C. gigas*	Atlantic	Bay	2011	187	39	148	20.8
Bourgneuf - Coupelasse	*C. gigas*	Atlantic	Bay	2014	200	48	152	24.1
La Fosse	*C. gigas*	Atlantic	Estuary	1987	370	68	302	18.4
La Fosse	*C. gigas*	Atlantic	Estuary	1990	220	49	171	22.3
La Fosse	*C. gigas*	Atlantic	Estuary	1993	350	66	284	18.8
La Fosse	*C. gigas*	Atlantic	Estuary	1996	270	56	214	20.6
La Fosse	*C. gigas*	Atlantic	Estuary	1999	280	35	245	12.4
La Fosse	*C. gigas*	Atlantic	Estuary	2002	230	41	189	17.7
La Fosse	*C. gigas*	Atlantic	Estuary	2005	120	22	98	18.5
La Fosse	*C. gigas*	Atlantic	Estuary	2008	290	32	258	11.0
La Fosse	*C. gigas*	Atlantic	Estuary	2011	234	36	198	15.3
La Fosse	*C. gigas*	Atlantic	Estuary	2014	350	48	302	13.6
Cap Ferret	*C. gigas*	Atlantic	Bay	1987	330	81	249	24.5
Cap Ferret	*C. gigas*	Atlantic	Bay	1990	240	49	191	20.3
Cap Ferret	*C. gigas*	Atlantic	Bay	1993	200	26	174	12.9
Cap Ferret	*C. gigas*	Atlantic	Bay	1996	230	47	183	20.6
Cap Ferret	*C. gigas*	Atlantic	Bay	1999	320	51	269	15.9
Cap Ferret	*C. gigas*	Atlantic	Bay	2002	160	42	118	26.1
Cap Ferret	*C. gigas*	Atlantic	Bay	2005	230	46	184	19.8
Cap Ferret	*C. gigas*	Atlantic	Bay	2008	100	30	70	30.2
Cap Ferret	*C. gigas*	Atlantic	Bay	2011	139	28	111	20.3
Cap Ferret	*C. gigas*	Atlantic	Bay	2014	250	51	199	20.5
Thau 4	*M. galloprovincialis*	Mediterranean	Lagoon	1987	100	53	47	53.5
Thau 4	*M. galloprovincialis*	Mediterranean	Lagoon	1990	140	63	77	45.1
Thau 4	*M. galloprovincialis*	Mediterranean	Lagoon	1993	100	34	66	34.4
Thau 4	*M. galloprovincialis*	Mediterranean	Lagoon	1996	130	45	85	34.8
Thau 4	*M. galloprovincialis*	Mediterranean	Lagoon	1999	90	50	40	55.8
Thau 4	*M. galloprovincialis*	Mediterranean	Lagoon	2002	100	57	43	56.6
Thau 4	*M. galloprovincialis*	Mediterranean	Lagoon	2005	100	43	57	42.6
Thau 4	*M. galloprovincialis*	Mediterranean	Lagoon	2008	120	53	67	44.5
Thau 4	*M. galloprovincialis*	Mediterranean	Lagoon	2011	118	35	83	29.3
Thau 4	*M. galloprovincialis*	Mediterranean	Lagoon	2014	100	33	67	32.8

Cap Couronne	*M. galloprovincialis*	Mediterranean	Coast	1987	390	119	271	30.5
Cap Couronne	*M. galloprovincialis*	Mediterranean	Coast	1990	160	45	115	28.2
Cap Couronne	*M. galloprovincialis*	Mediterranean	Coast	1993	280	70	210	24.8
Cap Couronne	*M. galloprovincialis*	Mediterranean	Coast	1999	300	55	245	18.2
Cap Couronne	*M. galloprovincialis*	Mediterranean	Coast	2002	220	79	141	36.0
Cap Couronne	*M. galloprovincialis*	Mediterranean	Coast	2005	270	85	185	31.6
Cap Couronne	*M. galloprovincialis*	Mediterranean	Coast	2008	200	55	145	27.4
Cap Couronne	*M. galloprovincialis*	Mediterranean	Coast	2011	295	74	221	25.2
Cap Couronne	*M. galloprovincialis*	Mediterranean	Coast	2014	290	77	213	26.7
Toulon - Lazaret	*M. galloprovincialis*	Mediterranean	Bay	1987	370	86	284	23.3
Toulon - Lazaret	*M. galloprovincialis*	Mediterranean	Bay	1990	440	50	390	11.4
Toulon - Lazaret	*M. galloprovincialis*	Mediterranean	Bay	1993	230	38	192	16.3
Toulon - Lazaret	*M. galloprovincialis*	Mediterranean	Bay	1996	320	43	277	13.6
Toulon - Lazaret	*M. galloprovincialis*	Mediterranean	Bay	1999	500	105	395	21.1
Toulon - Lazaret	*M. galloprovincialis*	Mediterranean	Bay	2002	530	114	416	21.4
Toulon - Lazaret	*M. galloprovincialis*	Mediterranean	Bay	2005	540	172	368	31.8
Toulon - Lazaret	*M. galloprovincialis*	Mediterranean	Bay	2008	700	171	529	24.5
Toulon - Lazaret	*M. galloprovincialis*	Mediterranean	Bay	2011	520	68	452	13.2
Toulon - Lazaret	*M. galloprovincialis*	Mediterranean	Bay	2014	670	132	538	19.7
Golfe de la Napoule	*M. galloprovincialis*	Mediterranean	Coast	1987	250	69	181	27.5
Golfe de la Napoule	*M. galloprovincialis*	Mediterranean	Coast	1990	160	54	106	33.9
Golfe de la Napoule	*M. galloprovincialis*	Mediterranean	Coast	1993	270	71	199	26.2
Golfe de la Napoule	*M. galloprovincialis*	Mediterranean	Coast	1999	210	74	136	35.2
Golfe de la Napoule	*M. galloprovincialis*	Mediterranean	Coast	2002	130	45	85	35.0
Golfe de la Napoule	*M. galloprovincialis*	Mediterranean	Coast	2005	160	40	120	25.1
Golfe de la Napoule	*M. galloprovincialis*	Mediterranean	Coast	2008	140	24	116	17.4
Golfe de la Napoule	*M. galloprovincialis*	Mediterranean	Coast	2011	121	30	91	24.5
Golfe de la Napoule	*M. galloprovincialis*	Mediterranean	Coast	2014	140	41	99	29.3