Supplemental material

Bivalve tissues as a recorder of multi-decadal global anthropogenic and climate-mediated change in coastal areas

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Detailed protocol of the 'ROCCH' monitoring sampling (up to date, year 2023)

The French National Monitoring Network "ROCCH" ("Réseau d'Observation de la Contamination CHimique", IFREMER, https://littoral.ifremer.fr/Reseaux-de-surveillance/Environnement/ROCCH-Reseau-d-Observation-de-la-Contamination-CHimique-du-littoral) samples bivalves as bioindicators of chemical contamination each year since the end of 1970s, at about 150 stations along French coastlines (the number of stations has changed since the start of the network as well as sampling frequency that was quarterly for the first twenty years of the network). Three different species are targeted, the Pacific oyster *Crassostrea gigas* and the blue mussels *Mytilus edulis* in the English Channel and Atlantic facades, and *Mytilus galloprovincialis* in the Mediterranean. The sampling protocol for bivalve mollusks of the ROCCH monitoring network has been designed to obtain samples with consistent and homogeneous characteristics for measuring chemical contaminants concentrations. Bivalves are collected alive at fixed points (maximum tolerance of 180 m around the initial point) away from known anthropogenic point discharges and in winter (currently mid-February, with a tolerance of one tide before and after the target date, meaning 6 weeks amplitude spreading from the end of January to the beginning of March). Bivalves are collected in wild beds or facilities, ensuring a minimum stay of 6 months on-site before sampling. The selected individuals are adults of a single species and uniform size (30 to 60 mm long for mussels, 90 to 140 mm long for oysters, i.e. 2 to 3 years old). A minimum of 50 mussels or 10 oysters is required to constitute a representative pooled sample.

Bivalves are then depurated for 18 to 26 hours in decanted seawater collected near the collection site. Once extracted from the shell, the whole bodies (i.e. total soft-tissues) of each individual are collected, drained for 30 minutes, crushed, and then freeze-dried. The resulting samples are stored in acid-treated glass containers, covered with calcined aluminum foil, and closed with a plastic lid. After chemical analysis of a sample aliquot, the containers are stored indefinitely, without an expiration period, at room temperature, protected from light, with dehumidifier control to minimize moisture pick-up.

Reference for the protocol: Grouhel Anne (2023). Prescriptions techniques pour l'échantillonnage de mollusques du réseau national d'observation des contaminants chimiques (ROCCH). RST- RBE/CCEM/ROCCH 23-02. https://doi.org/10.13155/97878

Complete list of station names used in this study

The 32 stations selected for the present study belongs to six sea regions: English Channel (EC, 9 stations), Western Brittany (WB, 4 stations), Northern Bay of Biscay (NB, 5 stations), Central Bay of Biscay (CB, 5 stations), Arcachon Lagoon (AL, 3 stations), Gulf of Lion (GL, 6 stations). Note that there is no station 17. The ROCCH station name and Mnemonic refers to the official denomination given to each station by the ROCCH monitoring network.

Sea region	Station name (this study)	ROCCH station name	ROCCH Mnemonic	Latitude	Longitude
English Channel	EC-1-AMBL	Ambleteuse	002-P-032	50.807367	1.595467
English Channel	EC-2-PSQT	Pointe de St Quentin	006-P-009	50.2808	1.528533
English Channel	EC-3-VARE	Varengeville	008-P-013	49.920769	0.982003
English Channel	EC-4-ANTI	Antifer - digue	010-P-014	49.649085	0.150327
English Channel	EC-5-CHLV	Cap de la Hève	010-P-055	49.507414	0.061995
English Channel	EC-6-VILL	Villerville	011-P-005	49.294071	-0.248005
English Channel	EC-7-OUIS	Ouistreham	010-P-120	49.404078	0.123666
English Channel	EC-8-PBES	Port en Bessin	013-P-001	49.351567	-0.753183
English Channel	EC-9-BDVG	Bdv Grandcamp ouest	014-P-007	49.386333	-1.101267
Western Brittany	WB-10-PLAN	Pen al Lann	034-P-001	48.665109	-3.8944
Western Brittany	WB-11-PASS	Le Passage (b)	039-P-007	48.391067	-4.384965
Western Brittany	WB-12-PERS	Persuel	039-P-093	48.293827	-4.55006
Western Brittany	WB-13-AULN	Aulne rive droite	039-P-124	48.281083	-4.260048
Northern Bay of Biscay	NB-14-GUIL	Le Guilvin	060-P-001	47.56765	-2.9338
Northern Bay of Biscay	NB-15-PEBE	Pen Bé	066-P-003	47.430664	-2.467999
Northern Bay of Biscay	NB-16-CHEM	Pointe de Chemoulin	070-P-102	47.234632	-2.297076
Northern Bay of Biscay	NB-18-BOUR	Bourgneuf - Coupelasse	071-P-065	47.012326	-2.022981
Northern Bay of Biscay	NB-19-NOIR	Noirmoutier - Gresse-loup	071-P-068	46.95066	-2.146303
Central Bay of Biscay	CB-20-RIVE	Rivedoux	076-P-032	46.163319	-1.27077
Central Bay of Biscay	CB-21-PALL	Les Palles	080-P-004	45.967477	-1.141439
Central Bay of Biscay	CB-22-BOYV	Boyardville	080-P-033	45.96381	-1.225941
Central Bay of Biscay	CB-23-PONT	Pontaillac	084-P-015	45.625137	-1.056097
Central Bay of Biscay	CB-24-FOSS	La Fosse	085-P-007	45.475635	-0.984592
Arcachon Lagoon	AL-25-FERR	Cap Ferret	087-P-013	44.643947	-1.241245
Arcachon Lagoon	AL-26-JACQ	Les Jacquets	088-P-067	44.722282	-1.194579
Arcachon Lagoon	AL-27-COMP	Comprian	088-P-069	44.683949	-1.084577
Gulf of Lion	GL-28-BANY	Banyuls - Labo Arago	094-P-008	42.480613	3.138878
Gulf of Lion	GL-29-HRLT	Embouchure de l'Hérault	095-P-026	43.275633	3.440536
Gulf of Lion	GL-30-STMM	Les Stes Maries de la mer	106-P-018	43.443979	4.420552
Gulf of Lion	GL-31-CART	Anse de Carteau	109-P-025	43.375649	4.875562
Gulf of Lion	GL-32-COUR	Cap Courronne	109-P-027	43.3239826	5.053900023
Gulf of Lion	GL-33-POME	Pomègues Est	111-P-002	43.267317	5.300573

Details on carbon stable isotope analysis

All isotope results are expressed using the conventional delta notation: δ^{13}Csample = [(Rsample/Rstandard) - 1], where R = $^{13}C/^{12}C$, with per mil deviation (‰) from international standards, Vienna Pee Dee belemnite. External (Caffeine IAEA600) and internal standards were analyzed as references every 10 samples, within each batch of samples.

Proxies of global environmental change

Public databases hereafter were used to retrieve proxies for global effect to calculate trends and compare with bivalves. We extracted monthly averages for hydro-climatic teleconnection indices and atmosphere CO_2 variables over the available time series (usually longer than our study period, see below), then we cut over the period Jan-1980 to Dec-2021. For each time series, in order to integrate a possible lag with bivalves, environmental proxies were recalculated as yearly averages based on the 12 months preceding bivalve sampling, e.g. for bivalves sampled in February 2021, we averaged the monthly index values from March 2020 to February 2021 included. This calculation was performed for teleconnection indices (AMO, NHT, AO, NAO, EAP) and atmospheric CO_2 concentrations. The atmospheric CO_2 $\delta^{13}C$ data were already averaged for calendar year (same for seawater $\delta^{13}C$-CO_2, see below), thus, we considered the previous year to compare with bivalves, e.g. $\delta^{13}C$-CO_2 data (atmosphere and sea water) from 2020 were compared with bivalves sampled in 2021.

Atlantic Multidecadal Oscillation (AMO): We selected the "AMO unsmoothed, short (1948 to present)" monthly averages dataset available at https://psl.noaa.gov/data/timeseries/AMO/

Northern Hemisphere Temperature anomalies (NHT): We selected the "Global Anomalies and Index Data" monthly averages dataset the Northern Hemisphere surface ocean over the period Jan-1880 to Dec-2021 available at https://www.ncei.noaa.gov/access/monitoring/global-temperature-anomalies#anomalies

Arctic Oscillation (AO): We selected the "Monthly mean AO index since January 1950" available at https://www.cpc.ncep.noaa.gov/products/precip/CWlink/daily_ao_index/ao.shtml

Northern Atlantic Oscillation (NAO): We selected the "Monthly North Atlantic Oscillation Index (PC-Based)" monthly average dataset from Jan-1899 to Dec-2021 available at https://climatedataguide.ucar.edu/climate-data/hurrell-north-atlantic-oscillation-nao-index-pc-based

East Atlantic Pattern (EAP): We selected the "Indices CPC_Indices NHTI EA: East Atlantic Pattern data" monthly averages dataset from Jan-1950 to Dec-2021 available at http://iridl.ldeo.columbia.edu/SOURCES/.Indices/.CPC_Indices/.NHTI/.EA/T+exch+table-+text+text+skipanyNaN+-table+.html

Atmospheric CO_2 concentrations (CO_{2atm}): We selected the "Globally averaged marine surface monthly mean data" from Jan-1979 to Dec-2021 available at https://gml.noaa.gov/ccgg/trends/gl_data.html (CO_2 concentrations are expressed as a mole fraction in dry air, micromol.mol^{-1}, abbreviated as ppm).

Atmospheric-CO_2 $\delta^{13}C$ ($\delta^{13}C$-CO_{2atm}): We used the "Global $\delta^{13}CO_2$" data from Graven et al., (2017) calculated as annual averages over the period 1850-2015 (Table S1 from Graven et al., 2017: Compiled records of carbon isotopes in atmospheric CO_2 for historical simulations in CMIP6). Data for the period 2015-2021 were calculated by linear extrapolation. ($\delta^{13}CO_2$ are expressed in per mil)

$\delta^{13}C$ of CO_2 dissolved in seawater ($\delta^{13}C$-CO_{2aq}): We calculated these values from atmospheric-CO_2 $\delta^{13}C$ as detailed below. Note that the difference in $\delta^{13}C$-CO_{2aq} between regions was only -0.018‰ (Table S1) which is equivalent to the analytical precision for bivalves samples (<0.15‰), therefore we used a global (average of all regions) value of $\delta^{13}C$-CO_{2aq} as a proxy to compare with bivalves-$\delta^{13}C$ in statistical tests.

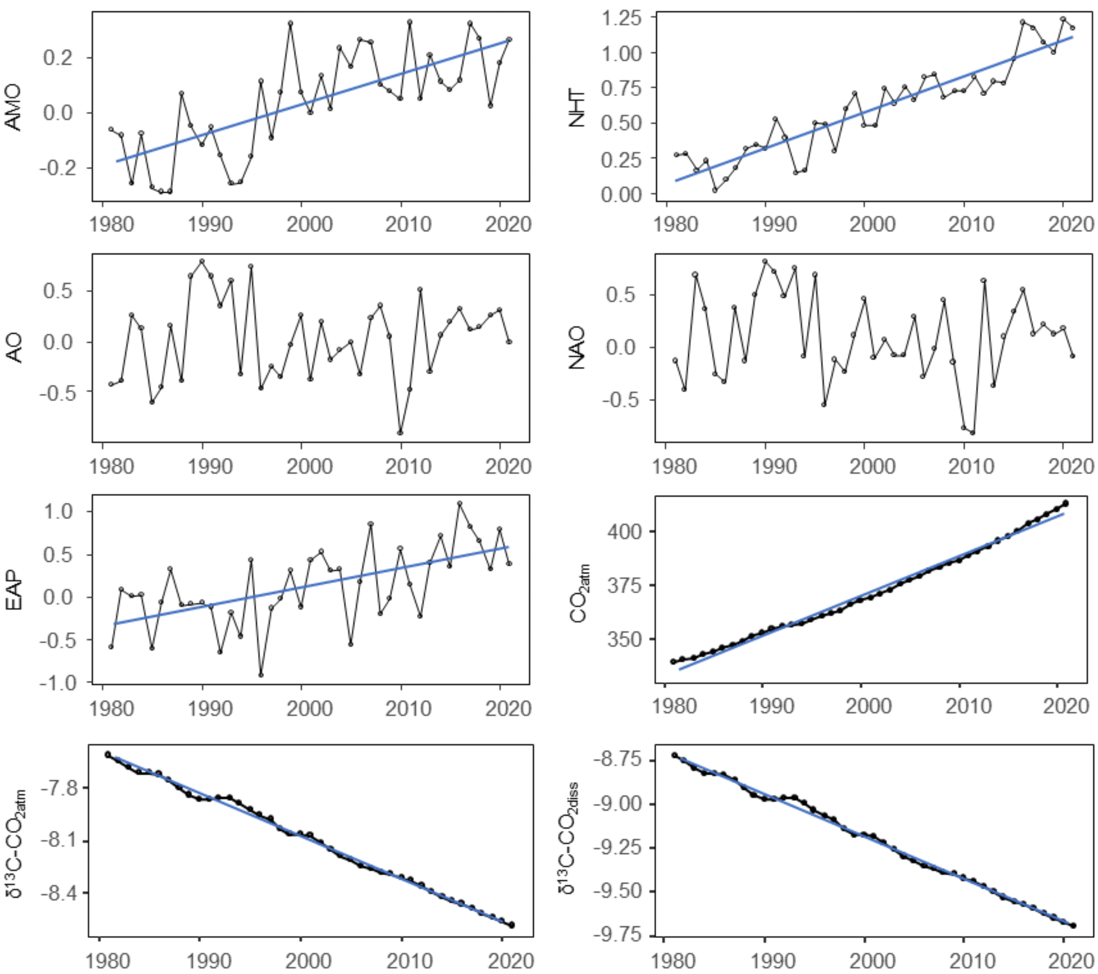

Figure S1: Pluri-decadal trends in hydro-climatic teleconnection indices (annual means, see indices names page 3), atmospheric CO_2 concentrations (ppm) and $\delta^{13}C$ of CO_2 in the atmosphere ($\delta^{13}C$-CO_{2atm} , ‰) and dissolved in seawater ($\delta^{13}C$-CO_{2aq}, ‰). Blue lines correspond to significant trends over the period 1981-2021 (Mann-Kendall tests; p-value < 0.05).

The Suess effect

The Suess effect was calculated from $\delta^{13}C$ of CO_2 dissolved in seawater ($\delta^{13}C$-CO_{2aq}) for each region, by adding the corresponding regional isotopic fractionation (ε) value to global $\delta^{13}C$-CO_{2atm} for each year on the period 1980-2021 (Table S1) as following:

The fractionation between gaseous and dissolved CO_2 phases was calculated from the equation of Vogel et al., (1970) as given by Mook (1986), and Zeebe and Wolf-Gladrow (2001): $\varepsilon d/g = -373/T + 0.19‰$ (T: absolute temperature in Kelvin). For each region, local temperature data were not always available for the entire time series and/or near bivalve stations, hence a regional mean temperature was calculated (average of all the time series of all the stations from a region ranging from 6 to 40 years, Table S1), which was used to calculate regional ε (average of all the time series of all the stations from a region, Table S1). Over time, we estimated the average bias resulting from the difference between the minimum and maximum ε values within each time series was only -0.009‰ (from -0.005 to -0.022‰ considering all time series together), hence, we considered ε as constant over time, neglecting any variations in temperature over the course of the time series. ε decreased with increasing temperatures at regional scale, from 1.115‰ at 12.7±0.7°C in the English Channel to -1.097‰ at 16.7±0.7°C in the Gulf of Lion (Table S1). Then, we calculated $\delta^{13}C$-CO_{2aq} for each region by adding regional ε values to global atmospheric-CO_2 $\delta^{13}C$ ($\delta^{13}C$-CO_{2atm}) over the period 1980-2021 (Table S1, standard deviation corresponds to multi-decadal variability in $\delta^{13}C-CO_{2aq}$ for each region). Maximum difference in $\delta^{13}C-CO_{2aq}$ between regions was only -0.018‰ which is lower than the analytical precision for bivalves samples (<0.15‰). Finally, the slope was calculated for each region (period 1980-2021) and gave the exact same value of -0.2431 ‰ decade^{-1} (the Suess effect, Table S1), showing a global Suess effect that was subtracted from bivalves. The Suess effect (slope) was removed from bivalve-$\delta^{13}C$ cumulatively for each year (starting from year 1981) to obtain Suess-corrected bivalve-$\delta^{13}C$ for each station.

Table S1: For each sea region, water temperature (average ± standard deviation calculated on all combinations of stations x years with data available), mean fractionation factor (ε; average ± standard deviation calculated on all combinations of stations x years with data available), $\delta^{13}C$ of CO_2 dissolved in seawater ($\delta^{13}C-CO_{2aq}$, average ± standard deviation calculated on the time series of each region over the period 1981-2021) and the decadal slope of $\delta^{13}C-CO_{2aq}$. Temperature data at stations the closest to bivalves sampling (when available) were retrieved from the REPHY monitoring program (https://www.ifremer.fr/surval) and recalculated as annual averages from the 12 months preceding bivalve sampling.

Region	Water temperature (°C)	Fractionation factor ε (‰)	$\delta^{13}C-CO_{2aq}$ (‰) 1980-2021	slope of $\delta^{13}C-CO_{2aq}$ (‰ decade^{-1})
English Channel (EC)	12.7 ± 0.7	-1.115 ± 0.003	-9.199 ± 0.295	-0.2431
Western Brittany (WB)	13.0 ± 0.5	-1.113 ± 0.002	-9.197 ± 0.295	-0.2431
Northern Bay of Biscay (NB)	13.7 ± 0.5	-1.110 ± 0.002	-9.194 ± 0.295	-0.2431
Central Bay of Biscay (CB)	14.3 ± 0.6	-1.108 ± 0.003	-9.191 ± 0.295	-0.2431
Arcachon Lagoon (AL)	15.4 ± 0.5	-1.103 ± 0.002	-9.187 ± 0.295	-0.2431
Gulf of Lion (GL)	16.7 ± 0.7	-1.097 ± 0.003	-9.181 ± 0.295	-0.2431

References

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