

Supplementary Information 1: Modern and archaeological site descriptions.

Mouchi et al.: 'Provenance study of oyster shells by LA-ICP-MS'

Localities of modern oysters

Baie des Veys (Normandy)

The Baie des Veys is located on the North side of the French coastline, in connection with the English Channel (**Figure 1-1**), which opens into the Atlantic Ocean. This area presents a semidiurnal tidal range of 8 m, and is characterized by an overall siltation induced by a poor resuspension of fine sediment particles caused by a weaker ebb than flow (Le Gall, 1970). This stronger flow limits the impact by the freshwater input from land on salinity in the bay (Sylvand, 1995). Two main channels recharge the bay with freshwater. The Carentan Channel, on its western part, is constituted by two main rivers, the Douve and the Taute, which drain a large part of the Bessin plain and the Cotentin. On the eastern part of the bay, the Isigny Channel is formed by the Vire and the Aure rivers. The watershed from these channels comprises limestone, basalt, acidic and alkaline metavolcanic rock and diorite (Baize et al., 1997).

Oysters collected from this locality are from a rearing experiment performed in a commercial oyster farm (Lartaud et al., 2010). The oyster farm is located several kilometres east from the Isigny Channel. During the experiment, shells were chemically labelled *in vivo* at a monthly rhythm, which allowed to specifically target the portions of the shells that were formed on site. Both *C. gigas* (n=5) and *O. edulis* (n=5) specimens were used in this study. The names of the groups are Cg_BdV_M and Oe_BdV_M, respectively.

Riec-sur-Bélon (Brittany)

Riec-sur-Bélon is located on the southern part of Brittany, on the Atlantic Ocean coastline (**Figure 1-3**). This locality is provided by freshwater from two river tributaries, namely the Aven and the Bélon. Tidal range is 5 m with a semidiurnal rhythm. The Bélon watershed is mainly composed of leucogranite, orthogneiss and micaschist rock (Béchenec et al., 1996, 2001; Monfort et al., 2006).

Historically, Riec-sur-Bélon was the first oyster farm to be implemented in Brittany during the 19th century. The oyster farm is located on the Bélon side of the estuary, where salinity is generally below 20 (Arzul and Quiniou, 2014). *Ostrea edulis* (n=3) were collected from this locality, and are referred to as Oe_RsB_M.

Marennes-Oléron (Charente-Maritime)

The Marennes-Oléron bay is located on the western part of France, on the Atlantic Ocean coastline (**Figure 1-4 A**). It opens on the Atlantic on its northern and southern borders by the pertuis of Antioche and the pertuis of Maumusson, respectively, while the Oléron Island borders the western part of the bay. A current from the Atlantic runs from North to South in the bay, and the tidal range is 5 m with a semidiurnal rhythm. The bay is recharged with freshwater from two rivers, the Charente (North side of the bay) and the Seudre (South side of the bay). The Charente has a rate of flow of 36 m³.s⁻¹, which represents 30 times that of the Seudre (Soletchnik et al., 1998). The Charente watershed (10,100 km²) crosses geologic formations of limestone and river deposits (Bourgueil et al., 1968; Bourgueil and Moreau, 1967, 1970; Platel et al., 1977, 1978) and originates from granite and gneiss formations in the Massif Central (Le Pochat et al., 1986). The Seudre watershed is substantially smaller, with only 780 km², and is composed of limestone and river deposits (Ternet and Berger, 1968; Platel et al., 1977, 1978).

Oysters are commercially farmed in the area, on the northern part of the bay. According to the direction of the Atlantic flow, this locality should be under the influence of the Charente river (and not the Seudre river). Specimens from *C. gigas* (n=3) and *O. edulis* (n=3) are referred to as Cg_MO_M and Oe_MO_M groups, respectively and were collected from a rearing experiment (Lartaud et al., 2010).

Tès (Arcachon basin)

The Arcachon basin is formed by a lagoon of 156 km² on the Atlantic Ocean coastline, on the western part of France, and 200 km south of the Marennes-Oléron bay (**Figure 1-5**). Oysters collected from this location were subject to a semidiurnal tidal range of 3 m. The basin is recharged with

freshwater from three main channels, the Eyre, the Porge and the Landes, as well as 26 other streams and ground water running through a watershed of 4,138 km². The watershed is composed mainly of Cenozoic river deposits as well as limestone and clay (Dubreuilh and Bouchet, 1992).

Only *C. gigas* specimens (n=8) were collected from this locality, and are designated as the Cg_Tes_M group.

Leucate (Aude)

The Salses-Leucate lagoon is a shallow coastal basin 14 km long and 5 km wide, located on the southwestern French Mediterranean coast (**Figure 1-6**). Freshwater supply is provided by groundwater discharges from karstic springs with a total flow of $5 \times 10^5 \text{ m}^3 \cdot \text{d}^{-1}$ (Fleury et al., 2007). Although the entire surface of the karstic waters is still unknown, it is estimated that it extends 60 km inland from the shore (Salvayre, 1989; Ladagnous and Le Bec, 1997). Mediterranean seawater penetrates the lagoon through three inlets separated by sandy barriers.

The Leucate lagoon also hosts a farming area, but the *C. gigas* specimens (n=5) presented here were collected from a natural broodstock living on the northern inlet of the pool. These specimens are designated here as Cg_Leu_M.

Diana lagoon (Corsica island)

The Diana lagoon (**Figure 1-7 A**) is located in the oriental coast of the island of Corsica (Tyrrhenian sea, NW Mediterranean). Its catchment area is ~62 km² and bounded to the west by relatively high shale massifs. To the east, it extends into the Tyrrhenian Sea coast by a continental shelf made up of Tertiary and Quaternary marine and alluvium formations. Quaternary deformations affected this coastal plain, which explains its hilly aspect and the formation of the Diana Lagoon (Casabianca 1967, Longère et al. 1972, Ottmann 1956).

The Diana lagoon stretches towards the southwest and extends over 550 to 570 ha (4 km in length and an average width of 2 km) with limestone, marls and sandstone (Caron et al., 1990). Connection with the sea was formerly carried out by a *grau* (local name for a channel connecting a

coastal pond with the sea) in the form of a 0.5 m deep gully, frequently silted up and therefore rarely open to the sea (Le Gall, 1953). Random connection with the sea would be the consequence of the low flow rates of watersheds, resulting in a slow renewal of water bodies outside the rare periods of opening of the *grau*. The water renewal rate would be relatively long, around 19 months (Paulmier 1996). According to Schachter and de Casabianca (1965), this *grau* was pierced by man. Currently work is regularly carried out to maintain a permanent opening in order to preserve shellfish and fish farms (Paulmier 1996, SOMIVAC – CTGREF, 1979).

Crassostrea gigas specimens (n=2) were collected from this site. The corresponding group is Cg_Dia_M.

Archaeological sites

Landévennec abbey (Brittany)

Landévennec (**Figure 1-2**) is located at the south of the Brest bay, on the last meander of the Aulne river (Bardel, 1991). In this archaeological site there is evidence of oyster exploitations from the 7th to 17th century. Two sites of flat oysters were sampled. The first one is an embankment on the exterior of a building dated to the 8-9th centuries CE (stratigraphic level 19611, n=1). The second one is a trench filling from the end of the 14th to the 15th century CE (stratigraphic level 19539, n=1).

Geology of the Aulne watershed comprises mainly schists, with sandstone, rhyolite and metagabbro (Babin and Darboux, 1982; Castaing, 1988; Le Gall, 1988; Plusquellec et al., 1999).

Ostrea edulis specimens collected from this site are referred to as the Oe_Lan_A group.

Brouage (Charente-Maritime)

The archaeological site of the Maison Champlain is inside the citadel port of Brouage opposite the Oléron Island between two rivers, the Charente and the Seudre (Champagne et al. 2012; **Figure 1-4 B**). During its construction in the 16th century CE, this citadel was in the middle of the Saintonge gulf opened to the ocean with marine influences. This gulf is currently filled and forms a dried marsh. The Brouage Channel skirted the citadel at the north of its walls. During the 16th and the 17th centuries,

ships were then sailing in this Channel, now partially filled by mud. The analyzed flat oysters are from the second half of the 16th century CE (stratigraphic level 3959). Due to the position of the site, it is supposed that the watershed corresponds to the Charente, described in the section above presenting the modern Marennnes-Oléron locality.

Oyster specimens (*O. edulis*; n=4) from this site are referred to as the Oe_Bro_A group.

Broue tower (Charente-Maritime)

The Medieval site of Broue is located on the North side of the Seudre river, on the Atlantic Ocean coast of France, in Charente-Maritime (**Figure 1-4 C**). The site is positioned on a peninsula surrounded by swamps fitted out as salt marshes. The site is composed of remains of a wall from a former church and a three-level, 15 by 28 m rectangular tower dated from the 11th century CE using dendrochronology and radiocarbon dating on charcoal (Normand et al., 2019).

The exploitation of local marine molluscs for human consumption is recognized from the 11th to the 15th c. CE. Like the Charente river for the Brouage locality, the exact position of the Seudre river during the 15th c. CE is unknown. Due to the position of the Tower however, it is possible that the oyster specimens collected from this site may have grown under the influence of the Seudre river (through surroundings streams), contrary to those from Brouage, or a mix between the Seudre and the Charente rivers, and under the influence of their respective watersheds (see description in the section above presenting the modern Marennnes-Oléron locality).

Oyster specimens (*O. edulis*) were collected from two stratigraphic units of the site: 1192 (n=3) and 1297 (n=3), dated to the 11th and the 14th centuries CE, respectively (Normand et al., 2019). Specimens from both stratigraphic units are gathered as the Oe_BrT_A group.

Aléria (Corsica island)

The colony of Aléria (Alalia) was founded by the Phocaeans on the east coast of Corsica around 565 BCE (Jehasse and Jehasse 1997). Located on a hill in the Eastern Plain, the ancient city is in the immediate vicinity of the mouth of the Tavignano, the second river in Corsica, and the Diana

Lagoon (**Figure 1-7 B**). Alalia was taken by the Romans in 259 BCE and became Aléria. Its proximity to the Latin coast and the influence of Etruscan culture made Aléria the capital of Roman Corsica and a prosperous city, the remains of which today constitute a major archaeological site (Bergeret et al., 2015; Coutelas and Allegrini-Simonetti, 2017) with a forum, a courtroom, villas, shops, a temple, an amphitheatre, thermal baths and sewers. The Diana pond, which probably housed the old port of war (*Dianæ portus*), has been well known since Antiquity for its oysters, which were consumed locally and allegedly exported to Rome (Jehasse and Jehasse, 1997). At the fall of Rome, around the 5th century CE, the Vandals, who swept over Corsica, razed the city (Jehasse and Jehasse, 1997). The analyzed shells originate from a 15-cm thick deposit, which has been recognized over a distance of 3 m inside the Roman forum.

The Tavignano watershed comprises schists, granite, gneiss, metabasalt, and to a lesser extent marls and sandstone (Jauzin et al., 1976; Amaudric du Chaffaut et al., 1985; Caron et al., 1990; Rossi et al., 1994). *Ostrea edulis* specimens (n=2) from this site are referred to as the Oe_Ale_A group.

La Malène (Occitany)

This site comprises fortifications dated from the 6th century CE, located on top of a cliff (Schneider and Clément, 2012), approximately 120 km north of the Mediterranean coastline of France (**Figure 1**). The *castrum* is thought to have hosted high social status occupants, due to the presence of golden currency and silver nails (Schneider and Clément, 2012), and represents one of the last sites from the Antiquity with *O. edulis* oyster shells as remains of consumption by the elite. It is considered that these oysters were transported over relatively short distances, indicating the Mediterranean coastline as an origin, yet without precise locality (Bardot-Cambot and Forest, 2014). This assumption is confirmed by the observation (Forest, 2020 for Mureau, 2020) of some oyster shells attached to valves of *Flexopecten glaber*, a bivalve species endemic to the Mediterranean Sea. Oyster specimens (n=6) from this site are indicated as the Oe_Mal_A group.

Cybèle (Lyons city)

The city of Lyons (Auvergne-Rhône-Alpes, eastern France; **Figure 1**) is rich in Antiquity sites dating from the 1st century BCE. In the area of the Fourvière hill, a pit filled by food wastes was discovered in proximity of building remains tentatively attributed to a sanctuary to the goddess Cybèle. Among the pit wastes, approximately 200 *O. edulis* oyster shells dated from the 1st century CE were found, whose origin is still debated (Bardot-Cambot, 2013). Based on morphometric measurements and associated mollusc shells, two groups of oyster shells were identified, with one tentatively attributed to the Mediterranean coastline and the other to the Atlantic coastline (Bardot-Cambot, 2013). Specimens from this site, collected from both morphometric groups, are referred to as Oe_X1_A (n=6) and Oe_X2_A (n=8), respectively.

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Supplementary Information 2: Quality assurance - certified reference material

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A total of 22 measurements of the BCR-2 basalt reference material have been performed for this study to address the quality of the measurements by LA-ICP-MS. The values are indicated below.

Ti	2.2736	2.3324	2.3595	2.3476	2.2848	2.3003	2.2649	2.2891	2.2674	2.3602	2.3099	2.3023	2.2599	2.3762	2.7395	2.2984	2.7671	2.2959	2.2954	2.2796	2.2557	2.2994
Cr	16.5774	16.8700	17.7942	18.2726	18.8189	16.9895	16.6198	16.5918	16.4741	17.2286	16.4020	16.6167	16.4305	17.8385	16.2219	17.4146	16.5426	16.8858	16.2256	17.1177	16.6614	16.5631
Cu	16.6029	17.3238	18.2190	16.9732	15.7939	16.6900	14.4835	17.5775	17.1590	16.4407	15.9323	16.6613	16.6255	16.3726	15.9883	16.7814	16.0706	16.7942	16.1808	16.6688	16.4055	16.9166
Zn	139.3879	144.5496	141.1708	143.7813	143.9091	133.3415	127.5495	137.7744	134.5128	131.5537	131.4955	140.6195	143.7138	134.3885	137.4514	146.7883	142.2726	142.9469	137.8980	134.6058	149.6410	133.5366
Sr	329.2198	331.7063	330.9741	334.6362	339.9092	331.9276	323.4683	327.8998	335.3608	335.6301	329.6827	330.1760	335.8523	328.6288	330.4466	325.9773	331.7110	334.7126	328.5690	326.7636	330.1479	324.6989
Y	31.4587	32.3494	31.9576	32.5076	32.5670	32.4271	31.4186	32.4185	33.3097	33.5834	32.8959	33.0287	32.6357	31.5167	31.9106	32.2697	33.2090	33.1196	32.5903	31.5066	31.7899	31.0894
Ba	650.2420	668.1087	724.8344	699.1811	699.8398	694.2838	656.0428	688.9788	675.2198	709.3624	686.2008	696.0376	685.5269	661.8345	673.0266	659.7785	681.0933	695.7817	679.2944	697.3440	712.1397	675.8795
La	24.2957	25.3058	25.0690	25.5478	24.9511	24.7224	24.8380	24.8347	25.2556	25.7947	25.7741	25.7954	25.1247	24.4167	25.1572	24.8928	25.4779	25.7475	25.4751	24.5332	24.4126	24.0641
Ce	51.8448	54.3902	56.5603	55.2400	54.2901	52.9865	50.6877	52.2842	53.3296	52.8364	53.9721	54.3498	53.0642	53.6355	52.5953	53.1375	53.5574	54.0159	53.1211	54.7381	54.0940	52.9243
Pr	6.4161	6.6396	6.5903	6.6933	6.7186	6.5474	6.2842	6.3479	6.5324	6.5845	6.6712	6.6445	6.5356	6.7186	6.5284	6.4325	6.5323	6.6700	6.4597	6.4588	6.4564	6.3971
Nd	27.6150	28.2644	28.5236	29.2309	29.1816	28.4382	27.4694	28.1593	28.4758	29.5044	28.6128	28.6380	28.2635	28.7205	28.3555	27.5664	28.5524	28.5883	27.7396	27.9169	27.6081	27.2994
Sm	6.2436	6.3813	6.4659	6.3958	6.4288	6.3493	6.2223	6.3458	6.4419	6.6506	6.5165	6.4522	6.4366	6.1684	6.3377	6.1972	6.4339	6.5630	6.3497	6.2732	6.1950	6.1361
Pb	10.3056	10.6500	11.5045	11.6650	12.0327	11.2178	10.4480	10.4911	9.7574	10.1300	10.0130	10.3265	10.4820	10.3795	9.8529	10.4622	10.1013	10.3810	10.2045	10.1623	10.3890	10.0684
U	1.6562	1.7724	1.8848	1.8177	1.7968	1.7234	1.5747	1.7243	1.7570	1.7056	1.7361	1.7958	1.7683	1.7572	1.7273	1.8086	1.7145	1.7759	1.7146	1.7763	1.7369	1.7557

GeoReM preferred values (GeoReM: Jochum et al. 2005):

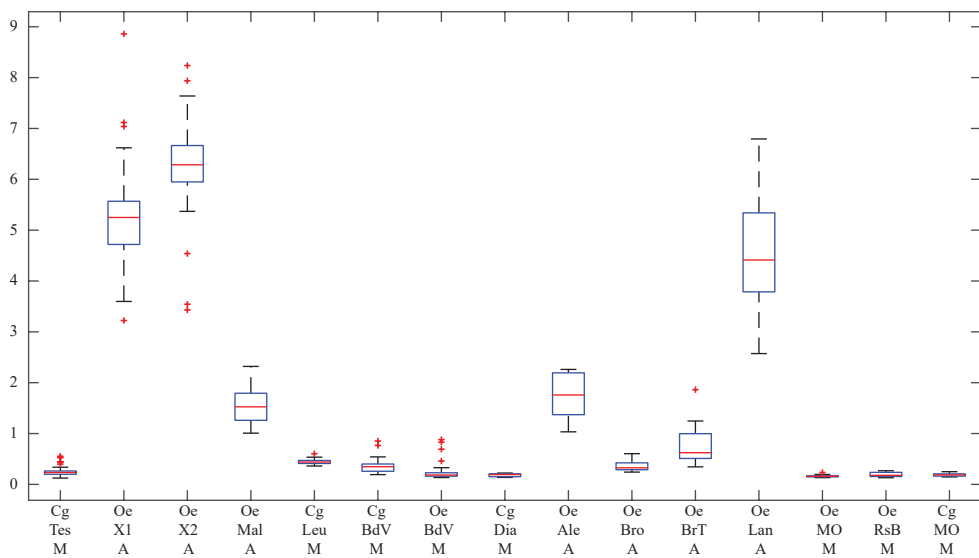
TiO₂	2.15 – 2.44 %m/m. 34 values (compiled: 2.26 – 2.265 %m/m. 3 values)
Cr	11 – 152 µg/g. 102 values (compiled: 15.85 – 18 µg/g. 3 values)
Cu	9.7 – 50.27 µg/g. 98 values (compiled: 19 – 19.66 µg/g. 2 values)
Zn	110 – 195 µg/g. 96 values (compiled: 127 – 129.5 µg/g. 3 values)
Sr	176 – 400 µg/g. 142 values (compiled: 337.4 – 346 µg/g. 3 values)
Y	28.13 – 52 µg/g. 127 values (compiled: 36.07 – 37 µg/g. 3 values)
Ba	445 – 808.96 µg/g. 128 values (compiled: 677 – 683.9 µg/g. 3 values)
La	20 – 31 µg/g. 135 values (compiled: 24.9 – 25.08 µg/g. 3 values)
Ce	46 – 66 µg/g. 136 values (compiled: 52.9 – 53.12 µg/g. 3 values)
Pr	6 – 15 µg/g. 132 values (compiled: 6.7 – 6.827 µg/g. 3 values)
Nd	25.5 – 37 µg/g. 165 values (compiled: 28 – 28.7 µg/g. 3 values)
Sm	5.9 – 10 µg/g. 161 values (compiled: 6.547 – 6.7 µg/g. 3 values)
Pb	7 – 18.1 µg/g. 121 values (compiled: 10.59 – 11 µg/g. 3 values)
U	1.46 – 2 µg/g. 165 values (compiled: 1.683 – 1.69 µg/g. 3 values)

Jochum, K.P. Nohl, U. Herwig, K. Lammel, E. Stoll, B. Hofmann, A.W. 2005. GeoReM: A new Geochemical database for reference materials and isotopic standards. *Geostand. Geoanalytical Res.* 29. 333-338. doi 10.1111/j.1751-908X.2005.tb00904.x.

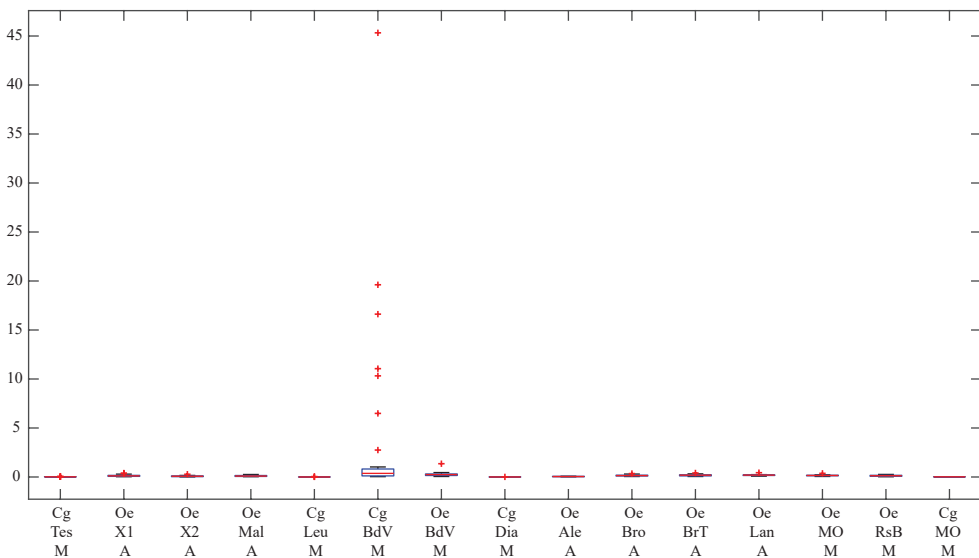
Supplementary Information 4
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shells by LA-ICP-MS'

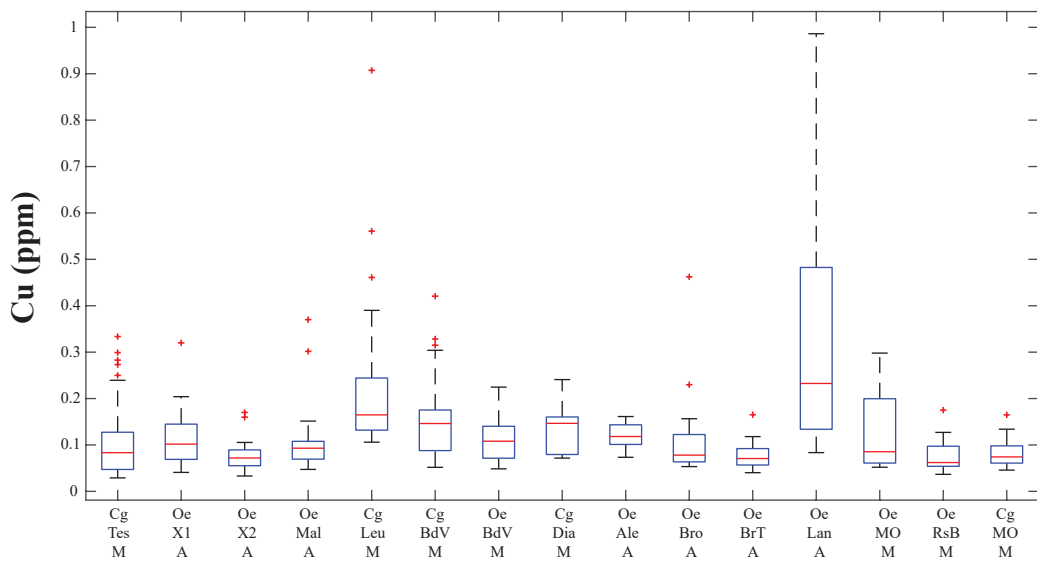
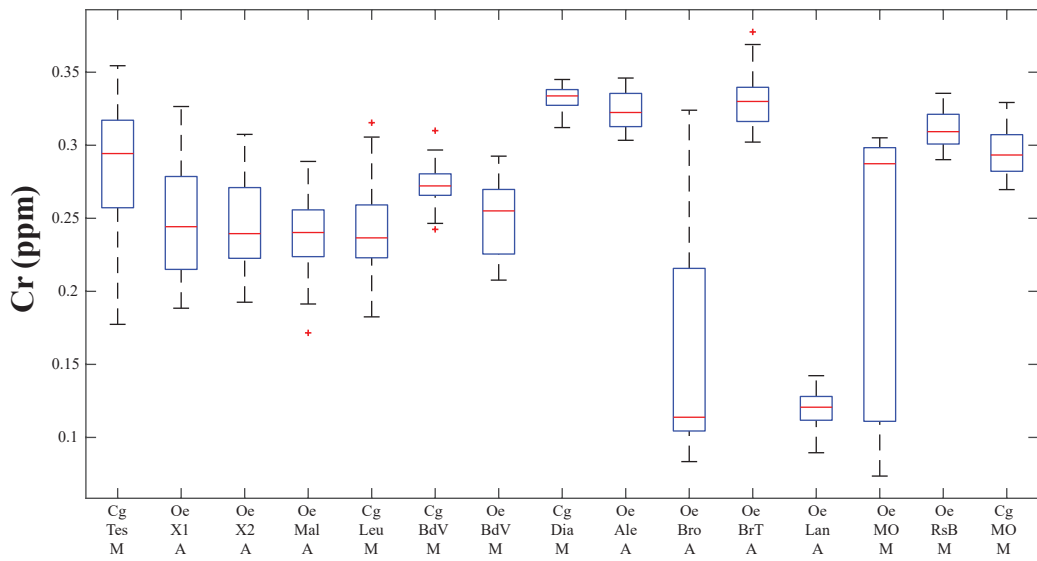
Boxplots by element for all shell groups.

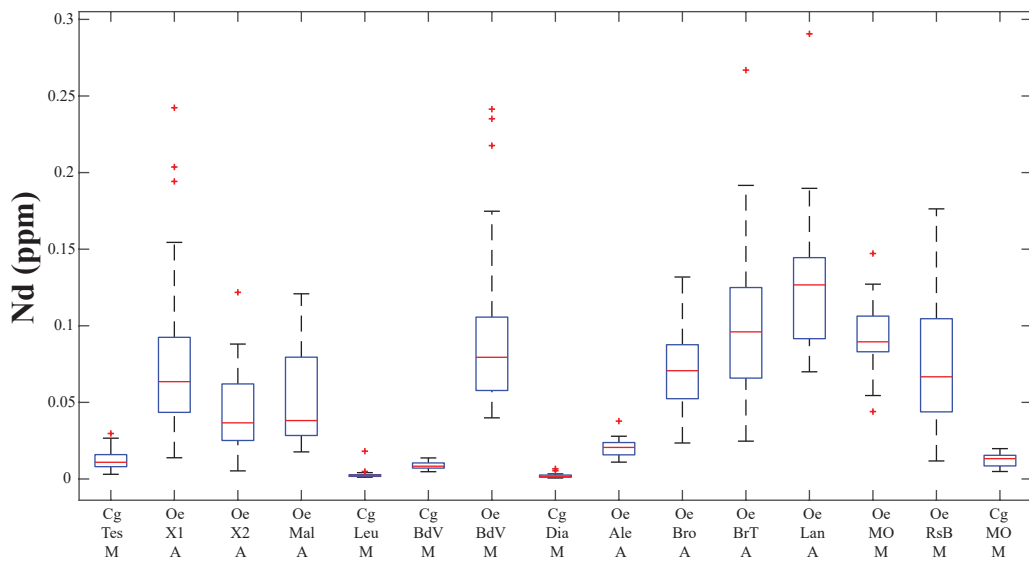
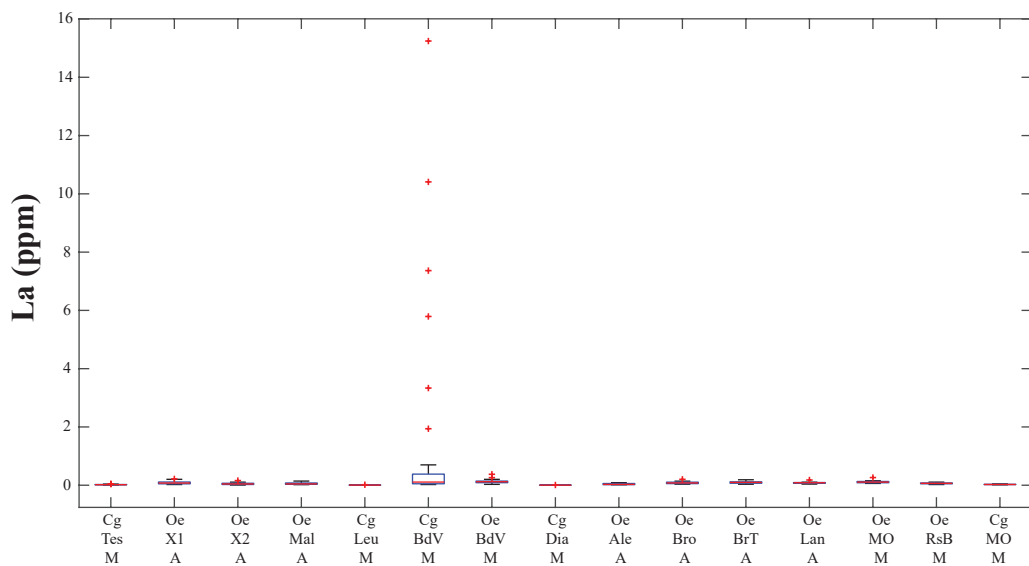
Ba (ppm)

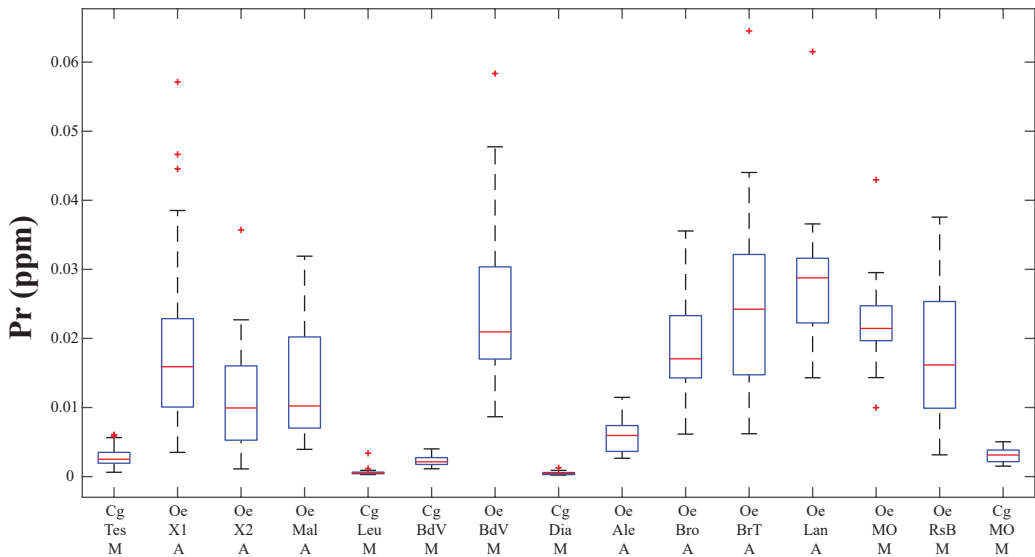
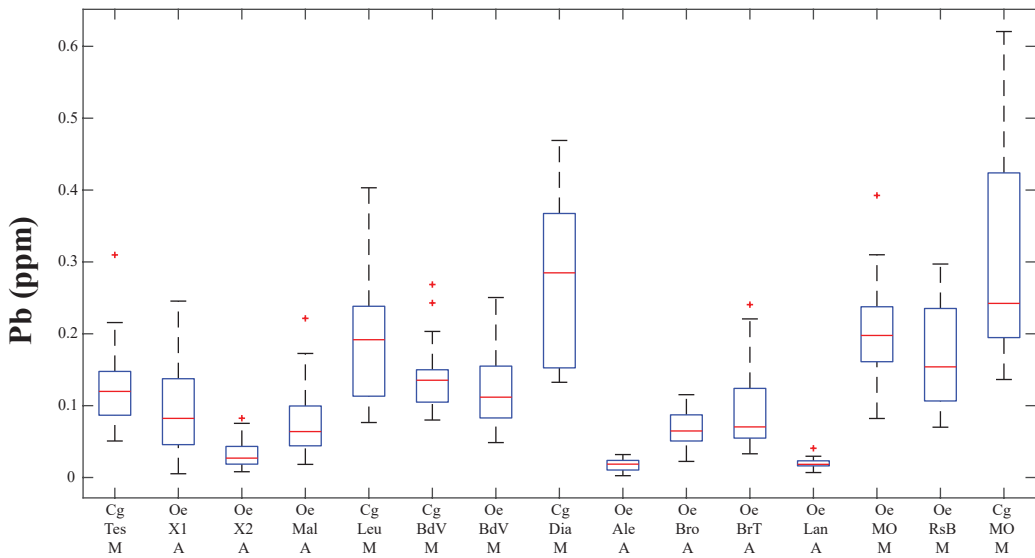


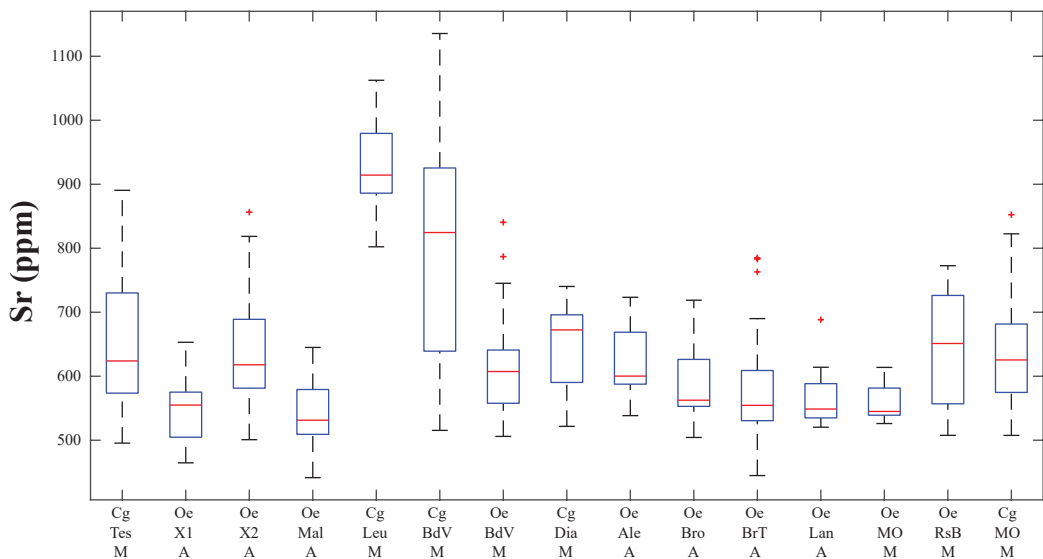
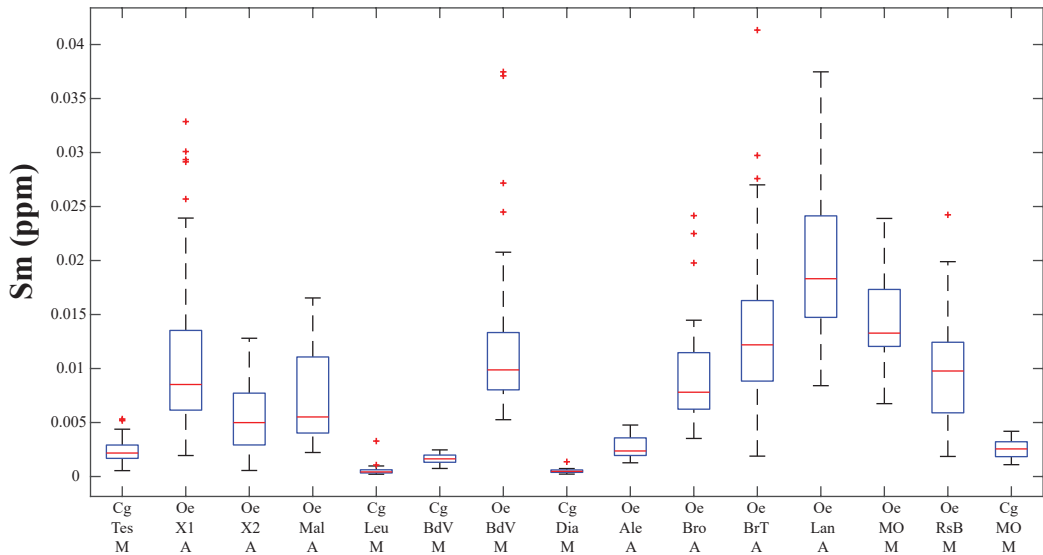
Ce (ppm)

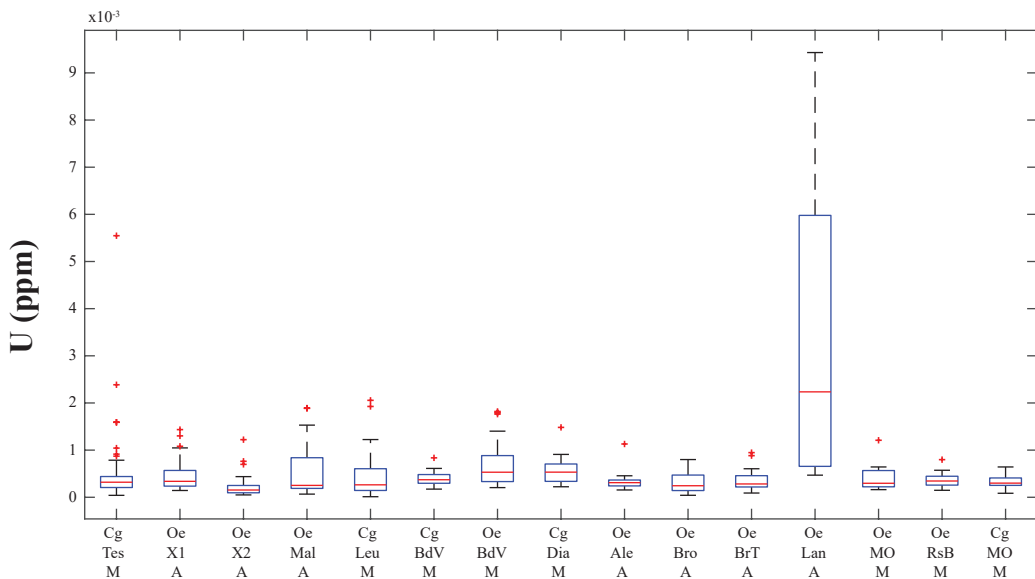
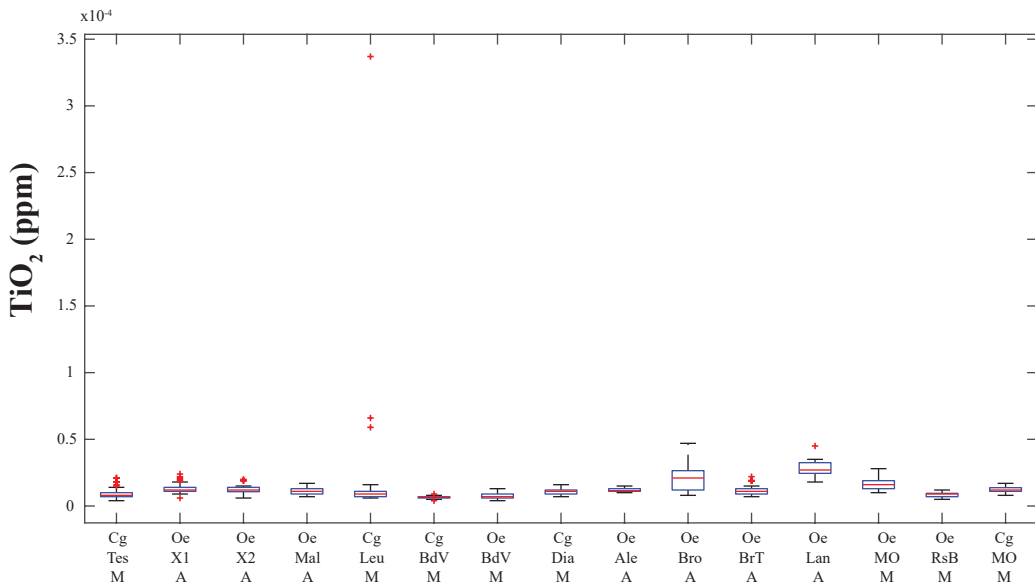


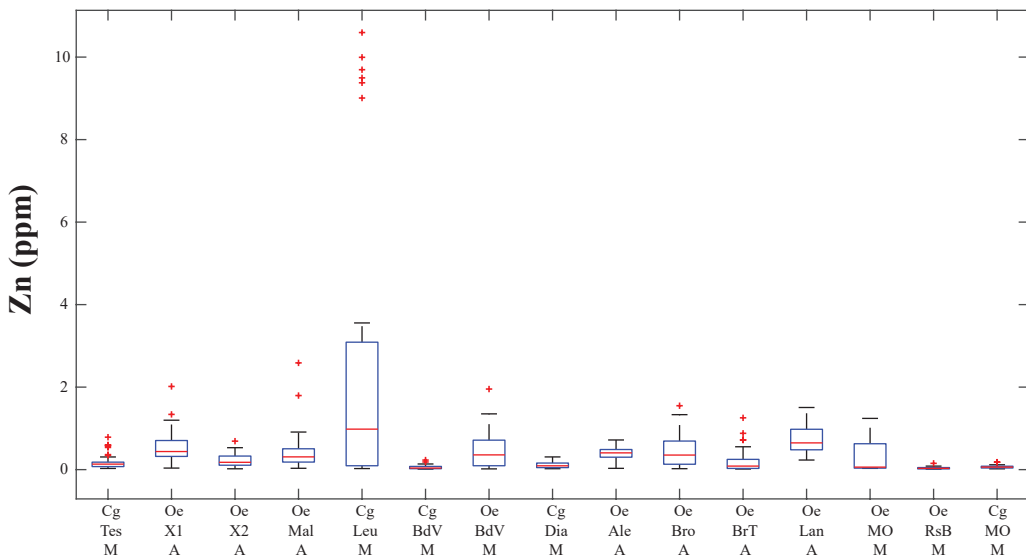
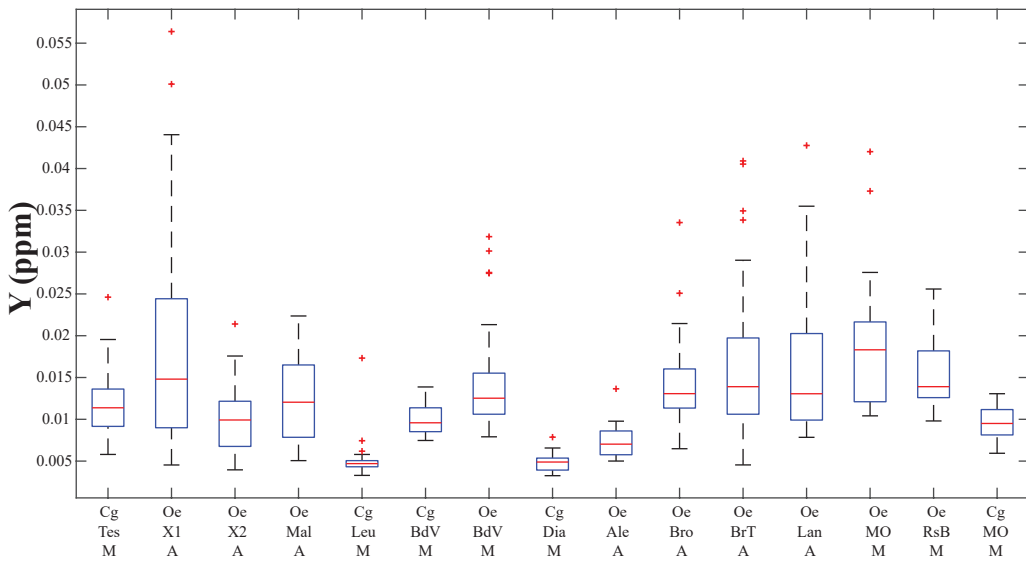










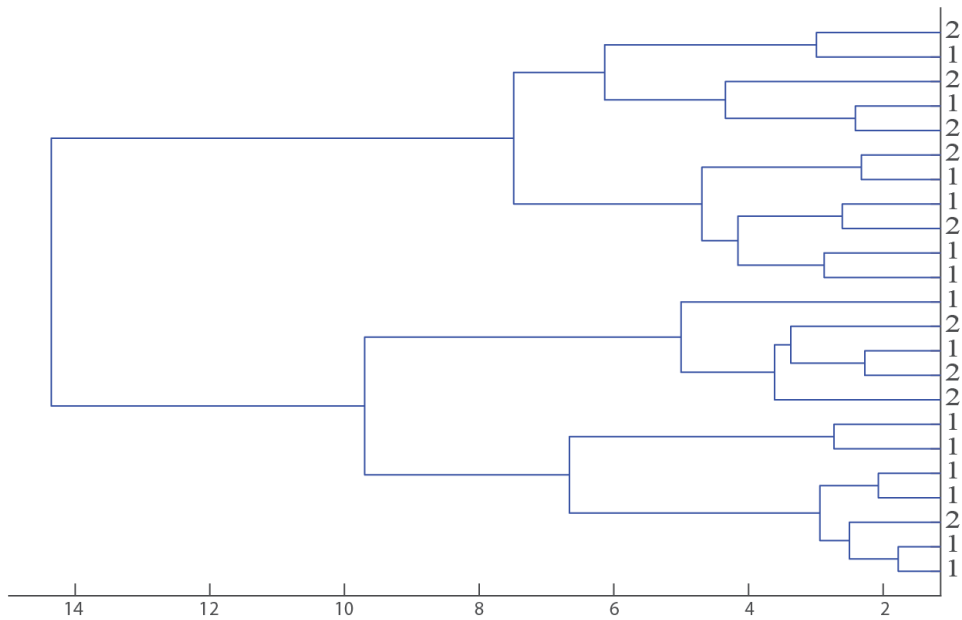


Supplementary Information 5: Seasonal influence of elemental composition of oyster shells.

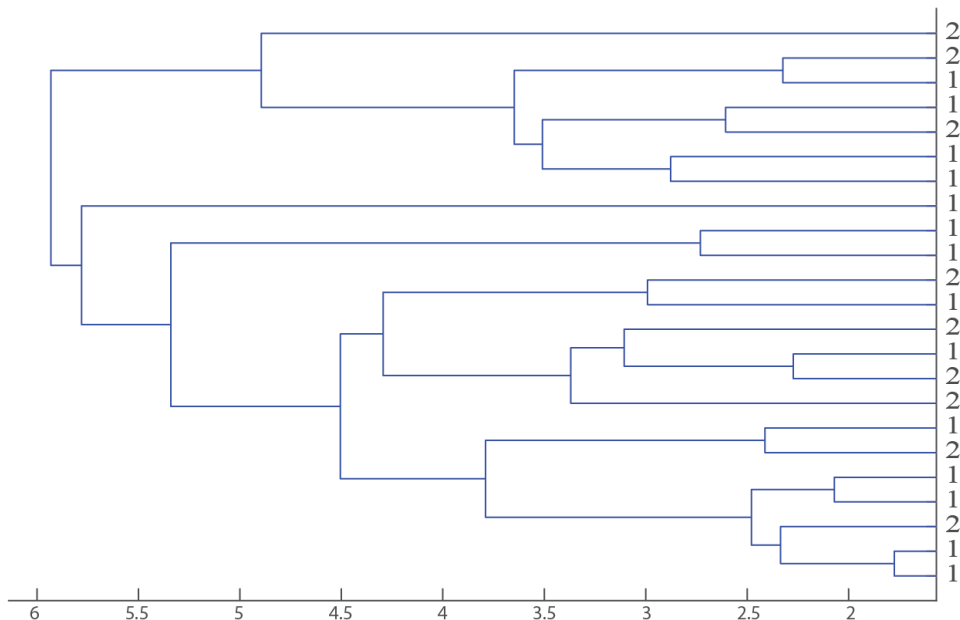
Mouchi et al.: 'Provenance study of oyster shells by LA-ICP-MS'

Dendrograms show that seasonal clusters are not supported by the elemental compositions of shells whatever the species (*C. gigas* and *O. edulis*) or the sites (Baie des Veys, Normandy, English Channel; and Marennes-Oléron, Charente-Maritime, Atlantic Ocean). Seasons are represented by '1' and '2'.

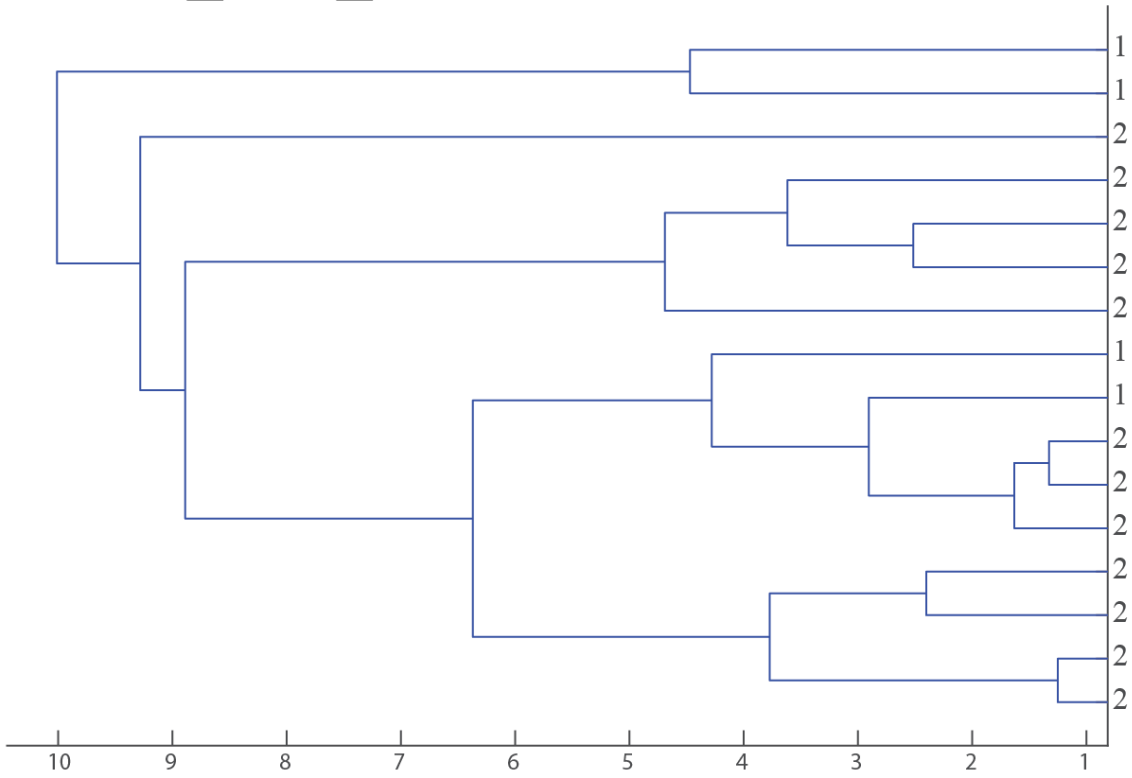
Cg_MO_M - Ward method



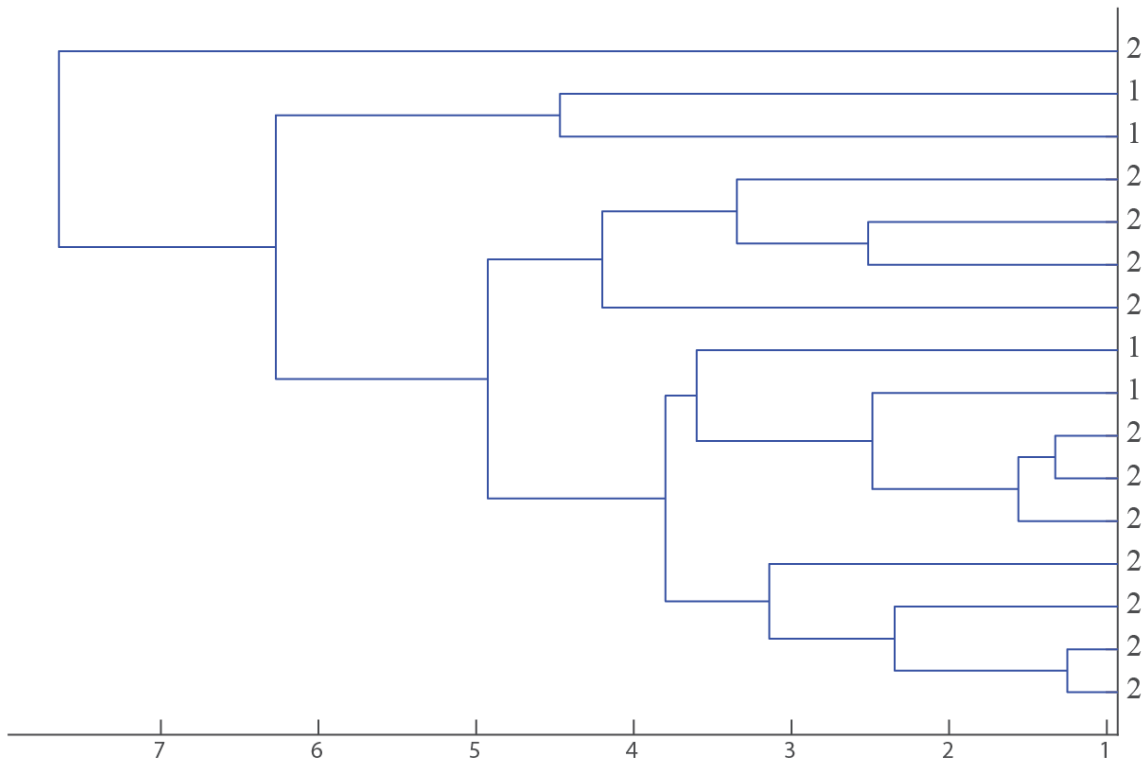
Cg_MO_M - Average method



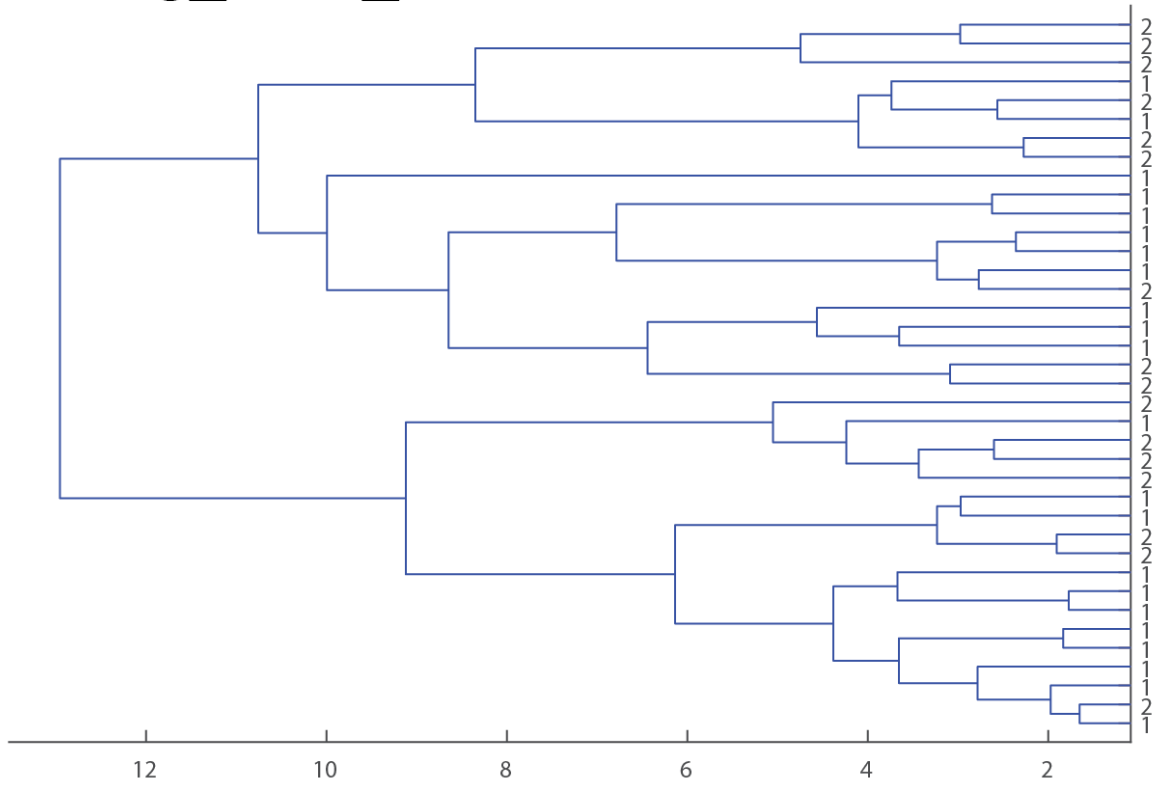
Oe_MO_M - Ward method



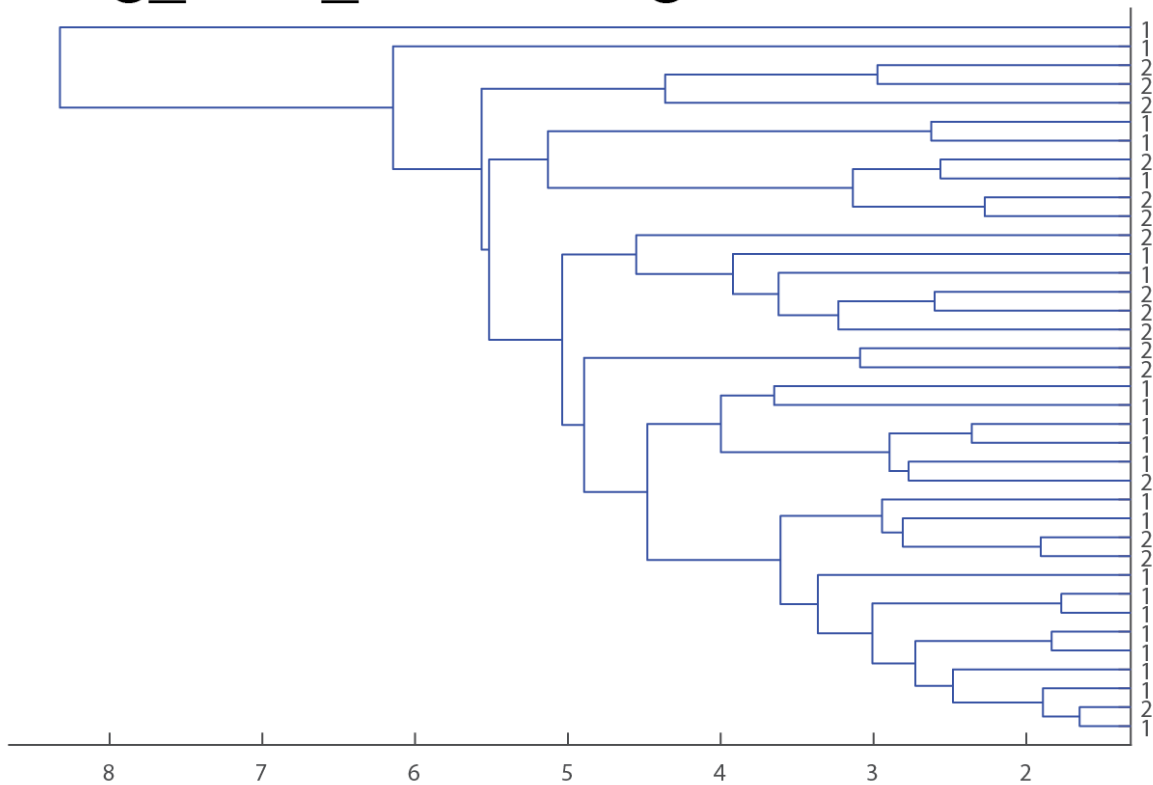
Oe_MO_M - Average method



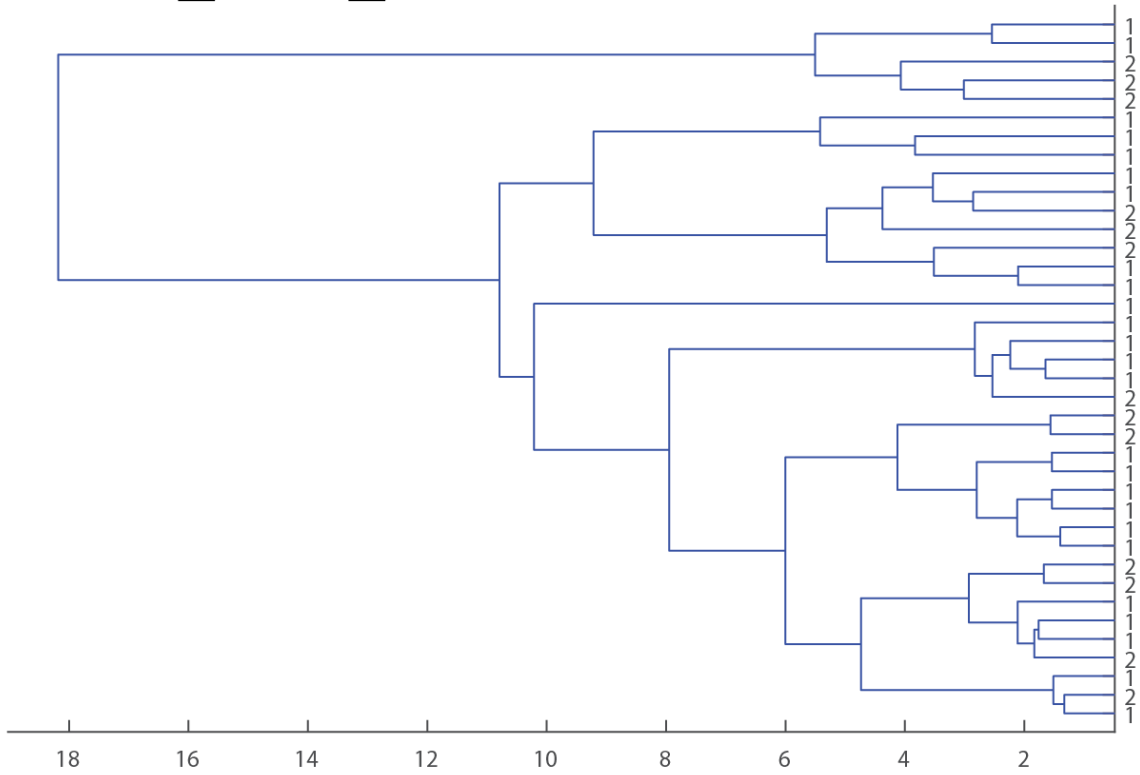
Cg_BdV_M - Ward method



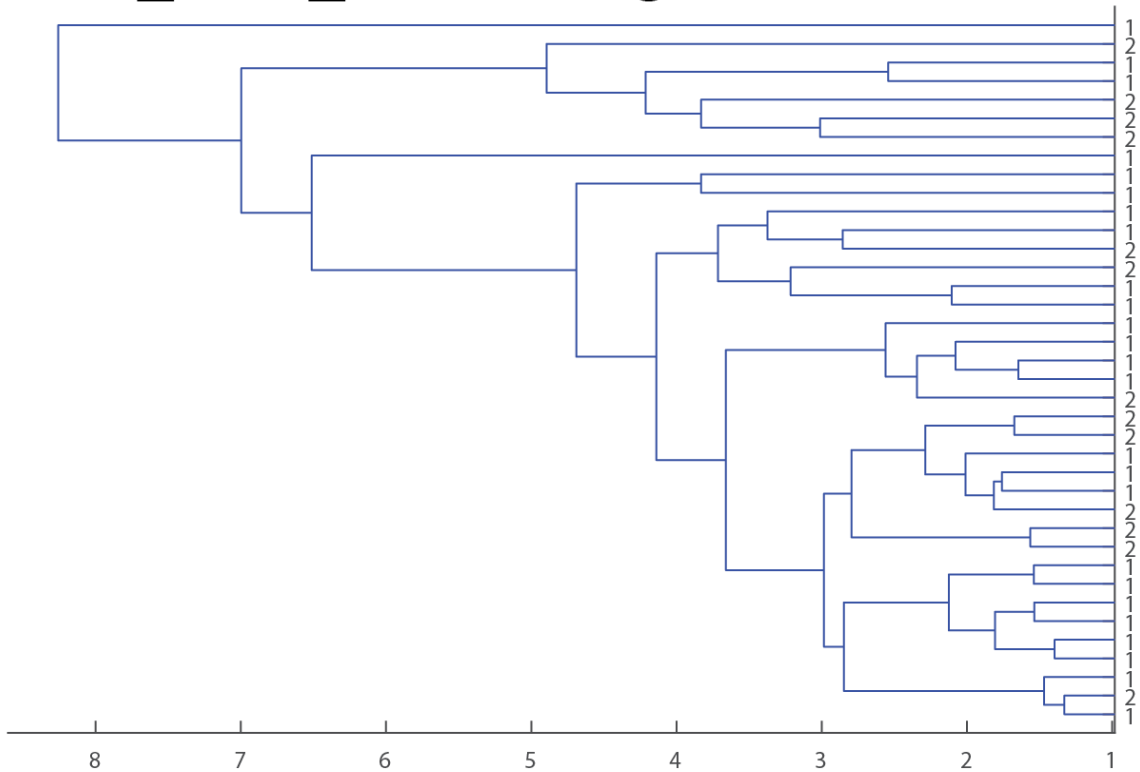
Cg_BdV_M - Average method



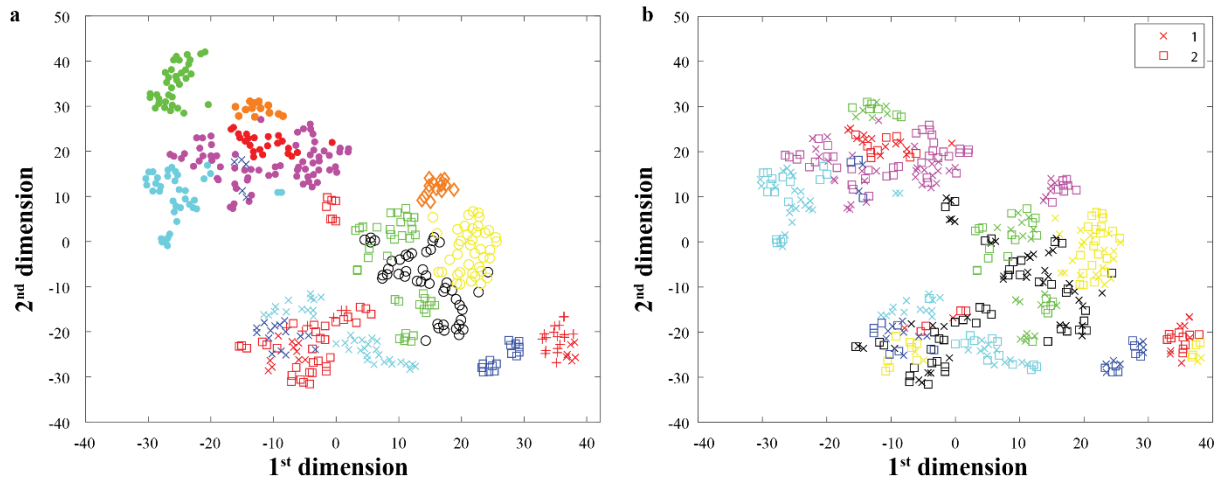
Oe_BdV_M - Ward method



Oe_BdV_M - Average method



Moreover, the distribution of all the measurements for the 14 groups of shells with seasonal calibration (*i.e.*, all the groups except Cg_Leu_M) visualized by t-SNE does not display significant differences between seasons, as indicated by the figure below.

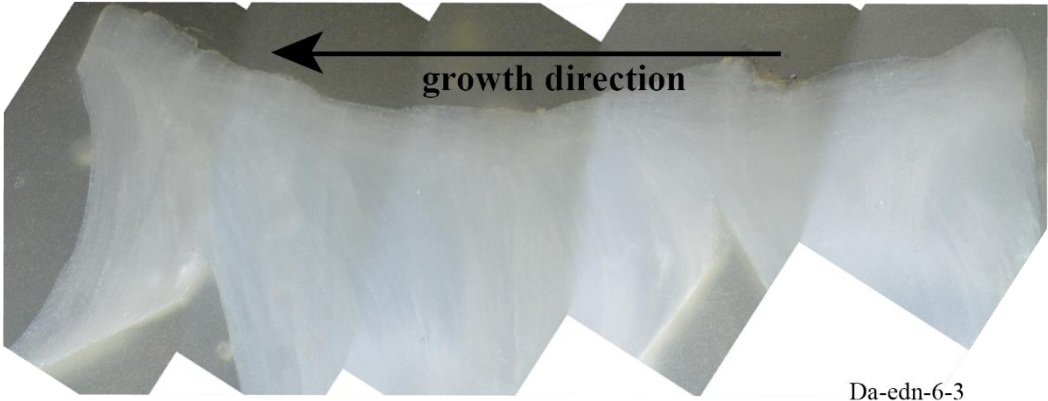


Supplementary Information 6: Temporal resolution of measurements from Modern labelled specimens from Marennes-Oléron (Oe_MO_M).

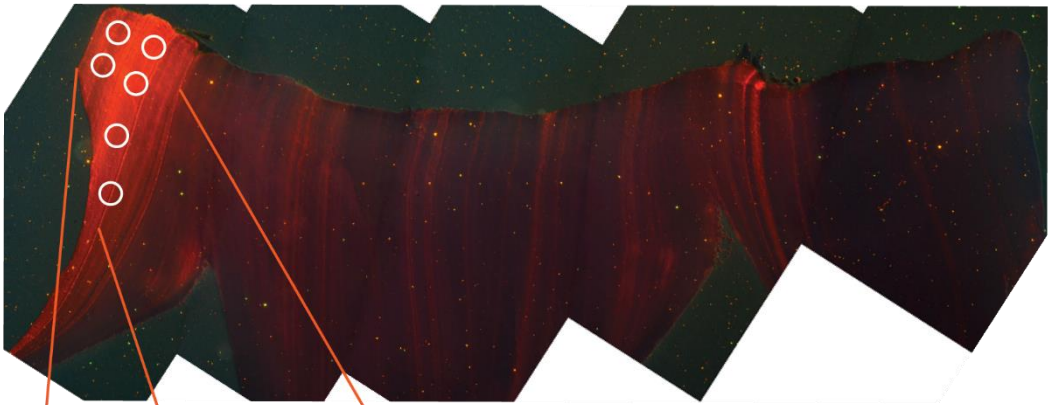
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The pictures below show natural light and cathodoluminescence observations of two *Ostrea edulis* specimens (Da-edn-6-3 and Da-edn-6-4) from a rearing experiment at Marennes-Oléron (Lartaud et al., 2010). These specimens were chemically labelled using manganese-enriched seawater on April 13th and June 12th 2006. The labels are visible by a bright growth increment in cathodoluminescence pictures. The position and size of the 200 µm measurements by LA-ICP-MS are indicated by white circles, and represent approximately half the space separating the two labels, which corresponds to one month of growth. Variations of this temporal resolution can occur according to shell growth rate during this period (*e.g.* reduced growth in winter months).

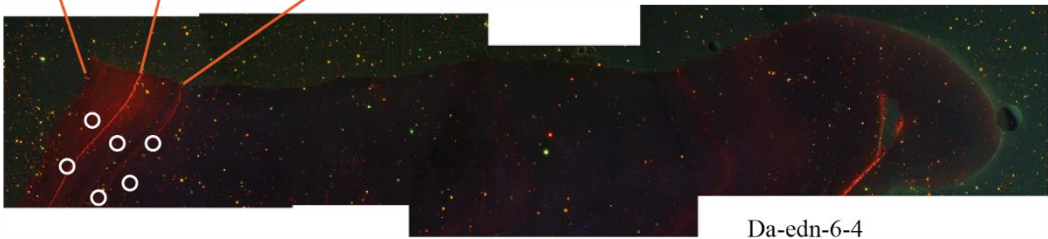
Lartaud, F., de Raféllis, M., Ropert, M., Emmanuel, L., Geairon, P., Renard, M., 2010. Mn labelling of living oysters: artificial and natural cathodoluminescence analyses as a tool for age and growth rate determination of *C. gigas* (Thunberg, 1793) shells. *Aquaculture*, 300 (1), 206-217, doi 10.1016/j.aquaculture.2009.12.018.



Da-edn-6-3



Nov. 22nd 2006 June 12th 2006 April 13th 2006



Da-edn-6-4



Supplementary Information 7: Influence of specific elements on discrimination.

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In our manuscript, we provide a method, using 14 elements, to discriminate the origin of multiple oyster shell groups from France and Corsica Island. These 14 variables appear to be the minimum to provide sufficient quality of the discriminating method. Here are provided t-SNE visualizations using the same dataset, with U, Ba and Pb respectively removed as variables. The visual distinction of groups from different origins is systematically reduced when removing a variable.

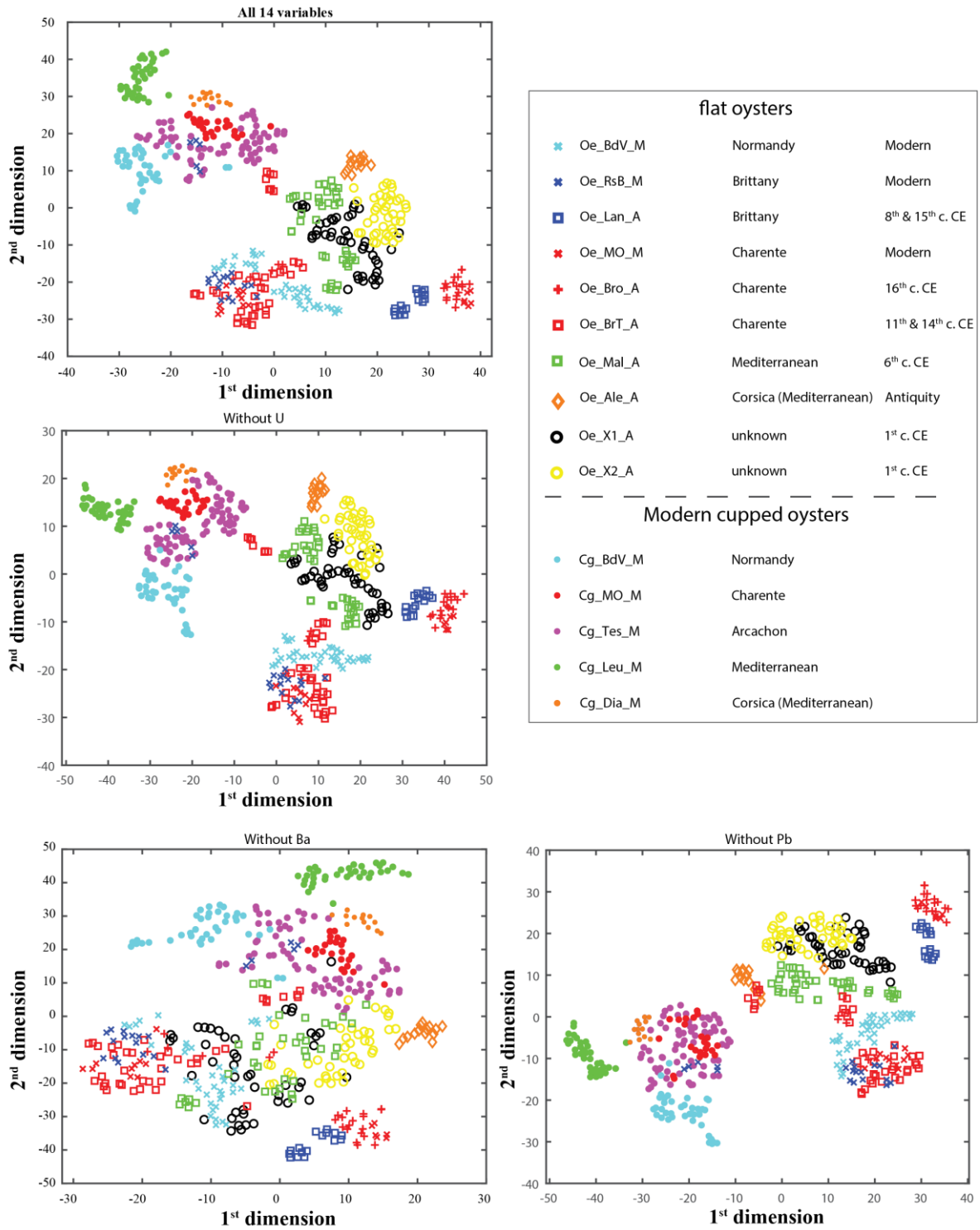


Figure above: Visualization of shell groups' partitioning using t-SNE showing discrimination of groups of shells being reduced when removing U, Ba and Pb from the variables.