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ABSTRACT

Provenance determination of archaeological remains is a valuable tool for reconstruction of past exchange networks. Among these materials, oyster shells are ubiquitous in sites from all

prehistorical and historical periods. Thus, they seem to be as promising candidates for provenance identification as they include chemical elements from the environment in their shells, which implies that an elemental fingerprint of the region of origin can be recorded in the shell composition. In this study, we present elemental measurements from 15 groups of modern and archaeological shells from 13 continental localities in mainland France and the island of Corsica (western Mediterranean Sea). Two of these localities had two oyster species (*Crassostrea gigas* and *Ostrea edulis*). Results indicate that (i) a species-specific elemental fingerprint exists and (ii) the Atlantic Ocean and Mediterranean Sea provenances can be identified for *O. edulis* shells. Moreover, if the shell originated from a locality only partially connected to the ocean (*e.g.* an estuary or lagoon), a fingerprint specific to the watershed can also be observed, even between groups originating from the same bay. Using these measurements as reference fingerprints, we characterize the Mediterranean origin of two groups of shells unearthed at Lyons (central France, 200 km away from the nearest shoreline), dated from the 1st c. CE.

1. INTRODUCTION

Reconstruction of exchange networks in archaeology is paramount to study large-scale medium- to long-term economics, as well as cultural and technological transfers, since routes were vectors of ideas and cultural exchanges. Numerous studies have attempted to reconstruct the provenance of marbles (Herz and Waelkens, 1988; Matthews, 1997; Mrozek-Wysocka, 2014), obsidian (Robin et al., 2016; Kuzmin et al., 2018), variscite (Querré et al., 2015), ceramics (Michelaki et al., 2013), and ore (Vogl et al., 2019). Yet, only a few studies have focused on shells and shell beads from chemical analyses (Shackleton and Renfrew, 1970; Eerkens et al., 2005).

In Mesolithic and Neolithic sites from the European Atlantic seaboard, food wastes composed of oyster shells are exclusively found on the coastline (Dupont, 2016) as the hunter-gatherer populations consumed the soft body and discarded the shell on site. More recent sites from Antiquity comprise oyster shells used for consumption in numerous localities, even distant from marine shores (Bardot-Cambot, 2014). Indeed, during this period, oysters were consumed by the elite as delicacies

away from the sea (Bardot-Cambot, 2014). Most of the time, the provenance of these archaeological shells remains unknown, unless these shells are associated with other taxa, (epi-)fauna, or substrate that presents some endemic characteristics possibly identified by specialists (Schneider and Lepetz, 2007; Bardot-Cambot, 2013; Chaufourier et al., 2015; Somerville et al., 2017). However, these identifications require substantial knowledge in cladistics as well as in the geological settings of the shoreline at regional to country (even sometimes continental) scales, as it is known that high-level dignitaries in Rome were used to import oysters from countries as far as England (Andrews, 1948; André, 1981). In the more common case of non-endemic associated items, archaeologists rely on morphometry to identify groups of shells of different origins (Gruet and Prigent, 1986a, 1986b; Bardot-Cambot, 2014). Nonetheless, such characteristics are not specific to a locality but rather, to the type of substrate (Gruet and Prigent, 1986a; Campbell, 2010; Somerville et al., 2017); therefore, they are not relevant to the geographical identification of the specimen's origin beyond a single region.

Other techniques have been developed in order to identify the provenance of shell remains, such as geochemical analyses, not only for archaeology (Eerkens et al., 2005, 2010; Mouchi et al., 2020a), but also for quality control of modern cultured specimens prior to market distribution (Bennion et al., 2019; Morrison et al., 2019). In addition, geochemistry has been used to study connectivity of separated populations of mollusc species that colonize distant localities through larval dispersion (Becker et al., 2005; Carson, 2010; Fodrie et al., 2011; Sorte et al., 2013).

Molluscs build their shells in calcium carbonate by sampling chemical elements from their surrounding waters, and the shell's chemical composition reflects (at least partially) that of the environment (Urey et al., 1951). Most studies have used oxygen and carbon stable isotope ratios ($\delta^{18}O$ and $\delta^{13}C$, respectively) from shells as a provenance proxy (Eerkens et al., 2005, 2010; Bajnóczi et al., 2013; Milano et al., 2019; Apolinarska and Kurzawska, 2020), but the use of this approach appears to be restricted to differentiate between marine and brackish/estuarine environments. Indeed, $\delta^{18}O$ and $\delta^{13}C$ are mainly representative of local biogeochemical conditions rather than the geographical location (temperature and salinity for $\delta^{18}O$ and food source for $\delta^{13}C$; Craig, 1965; Sharp, 2007; Lartaud et al., 2010a). Temporal homogeneity for the regional fingerprint cannot be insured as

seasonal temperatures have an impact on $\delta^{18}O$, leading to the overlap of amplitude values with remote areas exhibiting similar seasonal temperature contrasts (Mouchi et al., 2020b). Additionally, it was recently demonstrated that oysters do not mineralize their shell in equilibrium with seawater during their first year of growth, making unlikely the use of $\delta^{18}O$ as a proxy for local physicochemical conditions during this life stage (Huyghe et al., 2020). Overall, elemental analyses tend to provide better results in provenance determination than carbon and oxygen isotopic compositions (Zhao and Zhang, 2016; Ricardo et al., 2017; Bennion et al., 2019; Morrison et al., 2019). Nevertheless, such studies have identified issues related to 'vital effects': the metabolism acts as a filter between the living environment and the shell, thus modifying the shell's final composition. It has indeed been shown (Mouchi et al., 2020a) that concentrations in rare earth elements (REE) can be used to identify the region of provenance for the cupped oyster *Crassostrea gigas*. Still, these elements are not discriminant for the flat oyster, *Ostrea edulis*, which is the only oyster species found in European archaeological sites, as the cupped oyster *Crassostrea angulata* was imported to Portugal from Asia during the 17th century. Consequently, other chemical elements need to be investigated as provenance proxies for *O. edulis*.

In the present study, we use a new multi-elemental approach based on laser-ablation inductively-coupled plasma mass spectrometry (LA-ICP-MS) performed on 68 modern and archaeological oyster specimens (*C. gigas* and *O. edulis*) from 15 groups of different localities and ages. We aim at using this approach to identify potential discriminant elements for provenance studies applied to both oyster species found in Europe. This technique was selected as it is fast and commonly used in carbonate geochemistry studies (Durham et al., 2017).

2. MATERIALS AND METHODS

2.1. Collection sites and specimens

Samples were collected from multiple modern localities and archaeological sites in continental France and the island of Corsica (**Figure 1; Table 1**). As elemental composition of the rivers

(suggested to influence the composition of seawater and shells at the output) reflects the composition of substrate weathered on land, a brief description of the geological formations of watersheds are indicated with site descriptions in **Supplementary Information 1**. Specimens are divided into groups of origin, according to their species ('Cg' and 'Oe' for *C. gigas* and *O. edulis*, respectively), their locality (see **Table 1**), and their age ('M' for modern and 'A' for ancient specimens). Although the species *Crassostrea gigas* has been recently renamed *Magallana gigas* (Salvi and Mariottini, 2016), this genus change is debated by numerous experts (Bayne et al., 2017), so we will continue to use the name *Crassostrea gigas* throughout the paper.

Table 1: Number of specimens and measurements per group of shells.

Group	Location	Coordinates	ID on Figure 1	Age	Number of specimens	Number of measurements
Cg_BdV_M	Baie des Veys (Normandy)	49°23.11 N, 01°06.05 W	1	Modern	5	38
Oe_BdV_M	Baie des Veys (Normandy)	49°23.11 N, 01°06.05 W	1	Modern	5	38
Oe_Lan_A	Landévennec (Brittany)	48°17.42 N, 04°16.00 W	2	8^{th} – 15^{th} c. CE	2	16
Oe_RsB_M	Riec-sur-Bélon (Brittany)	47°48.55 N, 03°43.08 W	3	Modern	3	20
Cg_MO_M	Marennes-Oléron (Charente-Maritime)	45°52.23 N, 01°10.60 W	4A	Modern	3	23
Oe_MO_M	Marennes-Oléron (Charente-Maritime)	45°52.23 N, 01°10.60 W	4A	Modern	3	16
Oe_Bro_A	Brouage (Charente-Maritime)	55°85.05 N, 01°07.44 W	4B	16^{th} c. CE	4	24
Oe_BrT_A	Broue tower (Charente-Maritime)	45°47.67 N, 00°58.57 W	4C	11^{th} – 14^{th} c. CE	6	40
Cg_Tes_M	Tès (Arcachon Basin)	44°40.01 N, 01°08.18 W	5	Modern	8	70
Cg_Leu_M	Leucate (Aude)	42°52.48 N, 03°01.50 E	6	Modern	5	35
Cg_Dia_M	Diana lagoon (Corsica Island)	42°08.00 N, 09°32.17 E	7A	Modern	2	13
Oe_Ale_A	Aléria (Corsica Island)	42°06.50 N, 09°30.72 E	7B	Antiquity	2	14
Oe_X1_A	Unknown	Unknown	-	1^{st} c. CE	6	45
Oe_X2_A	Unknown	Unknown	-	1^{st} c. CE	8	41
Oe_Mal_A	Undefined (Mediterranean Sea)	Unknown	-	6^{th} c. CE	6	39

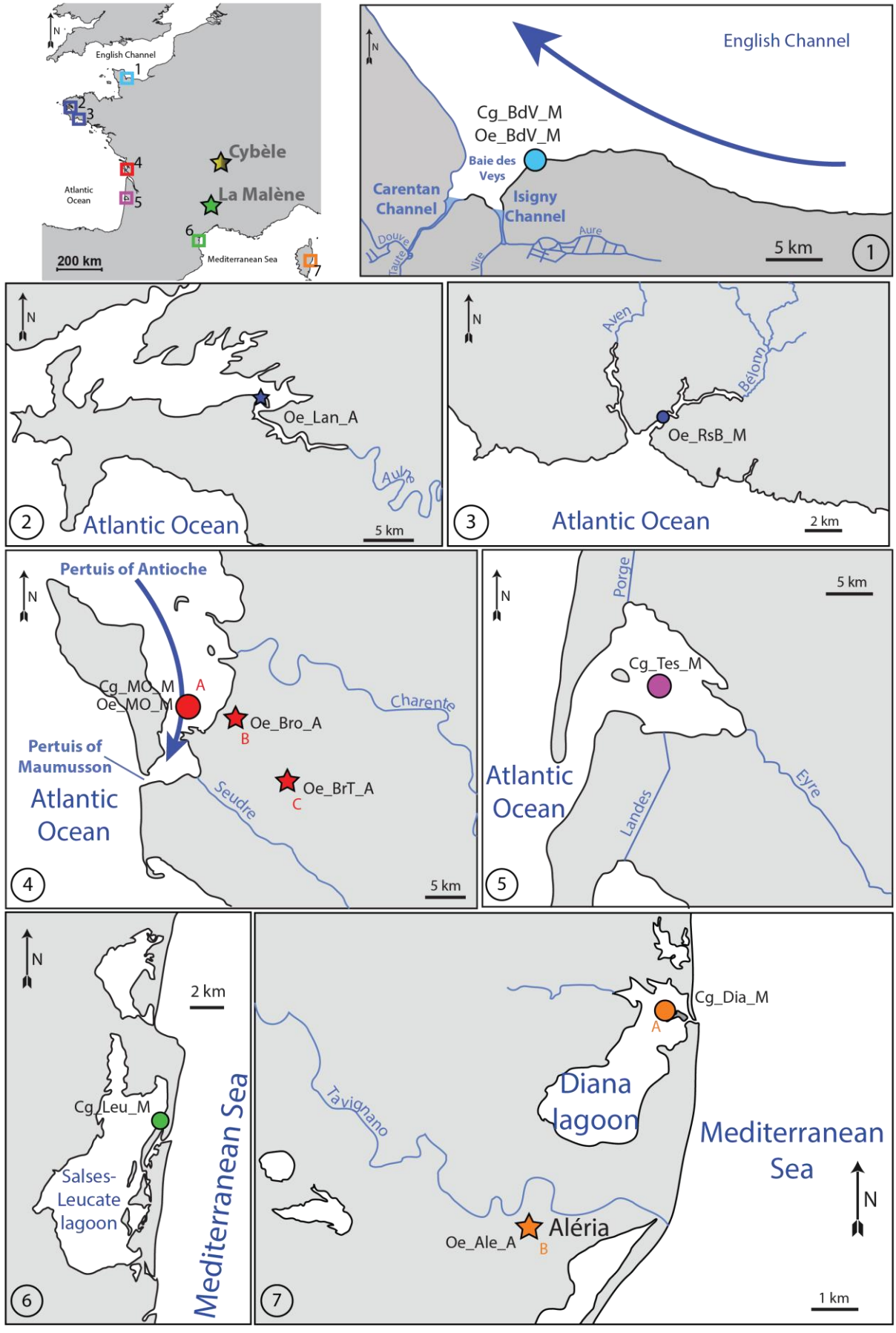

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Figure 1: Sample collection sites. Modern specimen groups are indicated by circles while archaeological specimen groups are presented as stars. Colour coding corresponds to that used in the t-SNE analysis on the following figures. 1: Baie des Veys modern site. Arrow: direction of the English Channel current near the shore (Lazure and Desmare, 2012). 2: Landévennec Medieval site. 3: Riec-sur-Bélon modern site. 4: Charente-Maritime, comprising the modern site of Marennes-Oléron (A), the Renaissance site of Brouage (B), and the Medieval site of Broue tower (C). Arrow: direction of the current in the bay (Dechambenoy et al., 1977). 5: Tès modern site. 6: Leucate modern site. 7: Corsica island sites, comprising the modern site of Diana lagoon (A) and the Antiquity site of Aléria (B). Additional archaeological groups are the Medieval shells found at La Malène of confirmed Mediterranean origin and two groups unearthed at Cybèle (Lyons) of unknown origin.

2.2. **Sample selection and preparation**

The preparation protocol is given in Mouchi et al. (2020a). Multiple specimens were selected from each group based on the apparent preservation of the umbo region of the shell. Epibionts, when present, were mechanically removed. The umbo of the left valve was embedded in Huntsman Araldite 2020 epoxy resin, cut through using a Buehler low-speed saw to expose the preserved internal structures along the growth axis, and made into polished 750 µm-thick sections. As the umbo records the entire growth of the shell in a condensed area, geochemical analyses on such sections have the benefit of being deprived of surface contaminations of chemical or organic origin (Richardson et al., 1993; Kirby et al., 1998). Indeed, the umbo is a dense area, protected from the exterior environment within the bivalve, which generally presents a much better preservation compared to the rest of the shell. In addition, oyster shells are made of lamellae that can be intertwined with empty spaces that can be filled with organic and sedimentary contaminants, which could strongly interfere with the quality of the measurements.

Each section was observed under cathodoluminescence to ensure the pristine state of the surface and to outline the seasonal calibration of the shell based on the intensity of luminescence (Lartaud et al., 2010b; Mouchi et al., 2018; Huygue et al., 2019). Following the growth direction, areas of strong luminescence correspond to parts of the shell built in summer periods, while areas in dull luminescence have grown during winter periods. Observations were completed using a Cathodyne-

OPEA cold cathode at ISTeP, Sorbonne Université (Paris, France) with 15-20 kV, 200-400 µA.mm^{-2} and a pressure of 0.05 Torr. Shells from Leucate (Cg_Leu_M) were the only specimens that did not exhibit cathodoluminescence, which made the seasonal calibration impossible. The seasonal calibration from morphology proposed by Kirby et al. (1998) was also unsuccessful due to the absence of the necessary curved surface of the umbo. All specimens from the other groups were seasonally calibrated.

2.3. Geochemical analyses by LA-ICP-MS

Measurements were performed using an Element XR (ThermoScientific) ICP-MS coupled with a RESOlution 193 nm ArF excimer ablation system equipped with an S155 two-volume ablation cell (Australian Scientific Instruments) at ISTE (University of Lausanne, Switzerland). The analyses were carried out with a pulse repetition rate of 20 Hz and an on-sample energy density of 4 J.cm^{-2}. Sample areas were cleaned from potential surface contamination (during polishing) by conducting pre-ablation. Spot size was 200 µm. Measurements were conducted on both summer and winter zones, based on the seasonal calibration. Measurements on Leucate specimens (Cg_Leu_M) were positioned randomly instead. Each specimen (shell) was measured several times in order to avoid bias due to internal variability. We carefully avoided performing measurements in suspicious areas regarding preservation, based on cathodoluminescence observation. We performed at least two adjacent measurements for each selected season area, with generally six to eight measurements per shell (sometimes less, *e.g.* four measurements for some Oe_MO_M specimens, due to lower growth rates restricting the surface available for measurements). For most specimens, three to four successive season areas were sampled. Prior and following each 15-sample series, NIST SRM 612 was measured for external calibration. Accuracy of the analyses was checked against measurements of the BCR-2 basalt reference material from USGS according to the GeoReM preferred values (Jochum et al., 2005). Relative standard deviation for all measured elements range from 1.2 % to 5.8 % (**Supplementary Information 2**). Measured elements were La, Ce, Pr, Nd, Sm, Ti, Cr, Cu, Zn, Sr, Ba, Pb, U, Y, and Ca as the internal standard. Data reduction was performed using the LAMTRACE software (Jackson, 2008). A total of 472 measurements were acquired.

2.4. **Data processing**

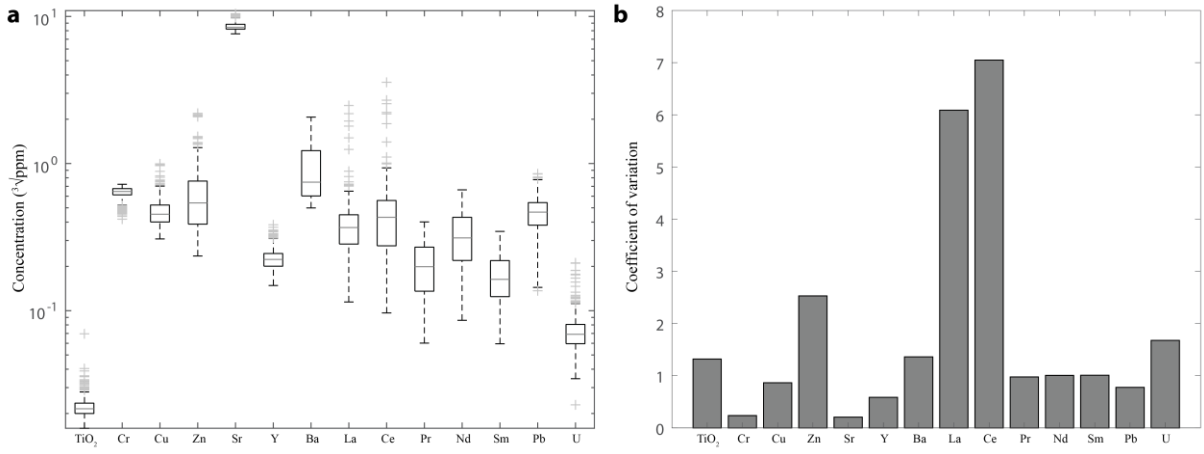
All data processing was conducted using the Matlab software (MathWorks, www.mathworks.com, v. R2017a). As none of the 14 measured elements presented a normal distribution (Kolmogorov-Smirnov test) due to a right-skewed distribution (towards larger element abundances), it was necessary to transform the data to normality for statistical processing, which was performed by using the cubic root transform (Chen and Deo, 2004). Hierarchical cluster analyses were used to estimate seasonal differences (using the seasonal age models based on the cathodoluminescence signal) at Baie des Veys on 38 measurements from *C. gigas* specimens (Cg_BdV_M; n=5) and 38 measurements from *O. edulis* specimens (Oe_BdV_M; n=5), and at Marennes-Oléron on 23 measurements from *C. gigas* specimens (Cg_MO_M; n=3) and 16 measurements from *O. edulis* specimens (Oe_MO_M; n=3). Two methods were tested: unweighted average distance and "Ward" inner squared distance. The cophenetic correlation coefficient between the distances obtained from the cluster tree and the original distances (in the multivariate space) is an indicator of the quality of the estimation (of the hierarchical cluster analysis) to faithfully represent dissimilarities between observations being compared. Comparison and classification between all groups were performed using t-SNE (t-distributed Stochastic Neighbour Embedding; van der Maaten and Hinton, 2008) with the exact Euclidean method. This method allows high-dimensional data points to be imbedded in low dimensions (typically two dimensions) in a way that respects similarities between points: distant points in a high-dimensional space correspond to distant embedded low-dimensional points, and nearby data points in the high-dimensional space correspond to nearby embedded low-dimensional points (MathWorks, www.mathworks.com, v. R2017a). In other words, each point represented by t-SNE corresponds to the overall composition of one measurement of all 14 elements, two close points having a similar composition while two distant points reflect different compositions. To prevent the undesirable over-influence of some elements showing wide variance, we performed t-SNE on normalized data. The Matlab script for t-SNE processing is available as **Supplementary Information 3**. As we focus on comparing the concentrations between individual shells from multiple localities using the same (LA-ICP-MS) analytical method, the host phase (organic

196 or mineral) of specific elements and their resulting co-variations within a measurement are not
197 discussed here.

198

199 ## 3. RESULTS

200 All measurement results are presented in **Figure 2a**. The three elements with the largest
201 coefficients of variation are Ce, La and Zn, followed by U, Ba, and Ti (**Fig. 2b**). All other elements
202 appear to have a low variance in the dataset. Except for Sr, Ba, Zn, La, and Ce, all measured elements
203 have concentrations below 1 ppm. Boxplots by element for all the shell groups are provided as
204 **Supplementary Information 4**. None of the elements can be used to differentiate between the origins
205 of the various groups on its own.



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207 **Figure 2:** Distribution of the 472 measurements for each analyte. **a:** Boxplot with values indicated in cubic root for best
208 approaching the normality in the distribution and for clarity. Note that the ordinate axis is in logscale. **b:** Coefficient of
209 variation for each analyte.

210

211 Similarities and differences between the 15 shell groups based on geochemical data are plotted
212 in **Figure 3** using t-SNE. First, the five groups of *C. gigas* shells are positioned at the upper left side of
213 the figure, while *O. edulis* samples are spread out in a much wider area. Moreover, the measurements
214 performed on both species from the same site (*i.e.*, Cg_BdV_M and Oe_BdV_M for Baie des Veys,
215 and Cg_MO_M and Oe_MO_M for Marennes-Oléron) do not overlap, indicating that the

compositions of the shells are not identical for both species from the same locality. Furthermore, the five groups of *C. gigas* shells are relatively well identified, particularly those of Leucate (Cg_Leu_M), Diana lagoon (Cg_Dia_M) and Baie des Veys (Cg_BdV_M). The Marennes-Oléron (Cg_MO_M) and Tès (Cg_Tes_M) groups partially overlap.

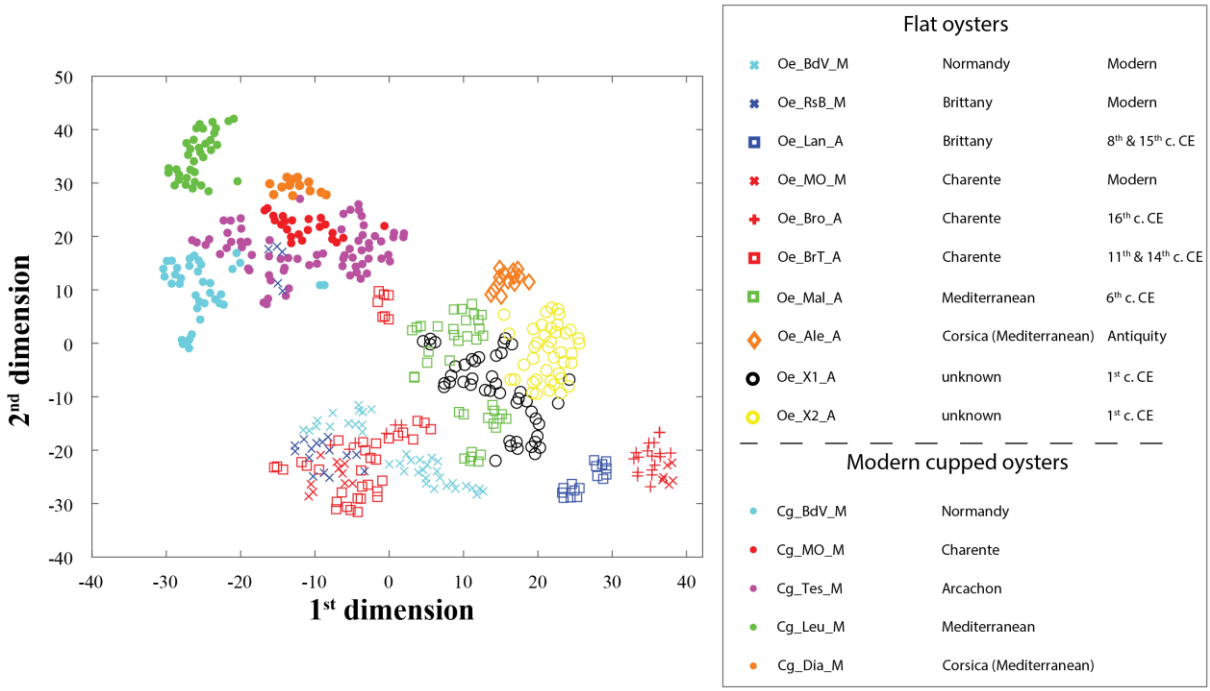

Figure 3: Visualization of shell groups' partitioning using t-SNE (exact method using Euclidean distance) based on 472 measurements. *Crassostrea gigas* shell measurements are indicated with dots. Light blue symbols represent measurements from shells originating from Normandy, dark blue symbols represent Brittany, red symbols represent Charente, purple symbols represent Arcachon Basin, and green and orange symbols represent the Mediterranean Sea, including Corsica. Two groups of Antiquity shells of unknown origin are indicated by yellow and black circles.

Although some *O. edulis* groups are well defined without any measurement overlap from other groups (*i.e.*, Oe_Ale_A and Oe_Lan_A), some groups of this species are more difficult to discriminate geochemically (**Figure 4a**). Indeed, when only considering groups originating from localities on the English Channel and the Atlantic Ocean coastlines (*i.e.*, open seas with strong tidal influence), numerous measurements cluster without discrimination between the locality of origin (dashed ellipse in **Figure 4b**). Only the Landévennec (Oe_Lan_A) group is absent from this common cluster. Inversely, the Oe_BdV_M group measurements are found solely in this common cluster. For the

Oe_RsB_M, Oe_MO_M, Oe_Bro_A and Oe_BrT_A groups, other measurements are found in 'secondary' clusters in addition to their presence in the common cluster, which are specific to the group. Some specimens from these groups have a unique fingerprint corresponding to one or the other cluster (common or secondary), while other specimens have fingerprints corresponding to both clusters along successive measurements, following shell growth (**Figure 5**). Amongst them, Oe_MO_M and Oe_Bro_A (both from Charente-Maritime) share this 'secondary cluster', while Broue tower (Oe_BrT_A; from the Charente-Maritime region as well) has a 'secondary cluster' isolated from the others (**Figure 4c**). It is also worth noting that specimens from the Oe_BrT_A group, which have been collected from two distinct temporal units in the site (see **Supplementary Information 1**), present measurements in the common cluster as well as in the secondary cluster for specimens from both stratigraphic units (sometimes along the same specimen), confirming that the local elemental fingerprint is identical over centuries. However, the two specimens from Landévennec (Oe_Lan_A), belonging to two different temporal periods, are discriminated from one another by t-SNE (**Figure 3**) in the form of two groups of measurements in the Landévennec cluster.

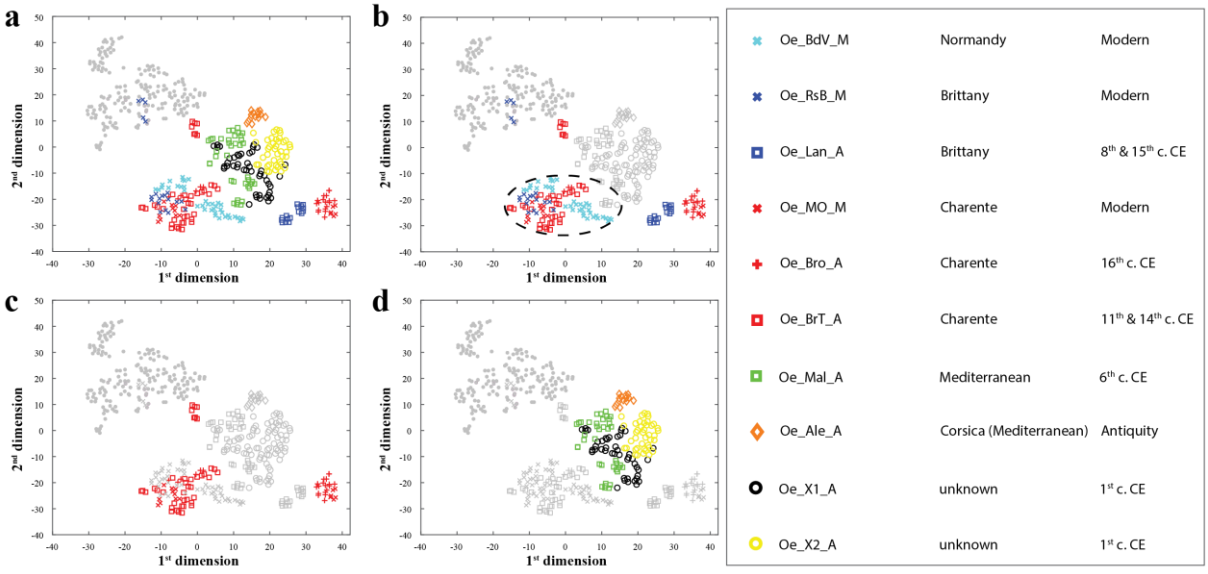

Figure 4: Visualization of *O. edulis* shell groups' partitioning using t-SNE showing specific groups. **a:** t-SNE from Figure 3 with measurements from *C. gigas* shells shaded. **b:** Same as **a** showing only groups of confirmed English Channel or Atlantic Ocean coastlines. The dashed ellipse indicates a cluster common to measurements from several groups. **c:** Same as **a** showing only the three Charente groups. **d:** Same as **a** showing only the groups from the Mediterranean coastline and the two groups of unknown origin.

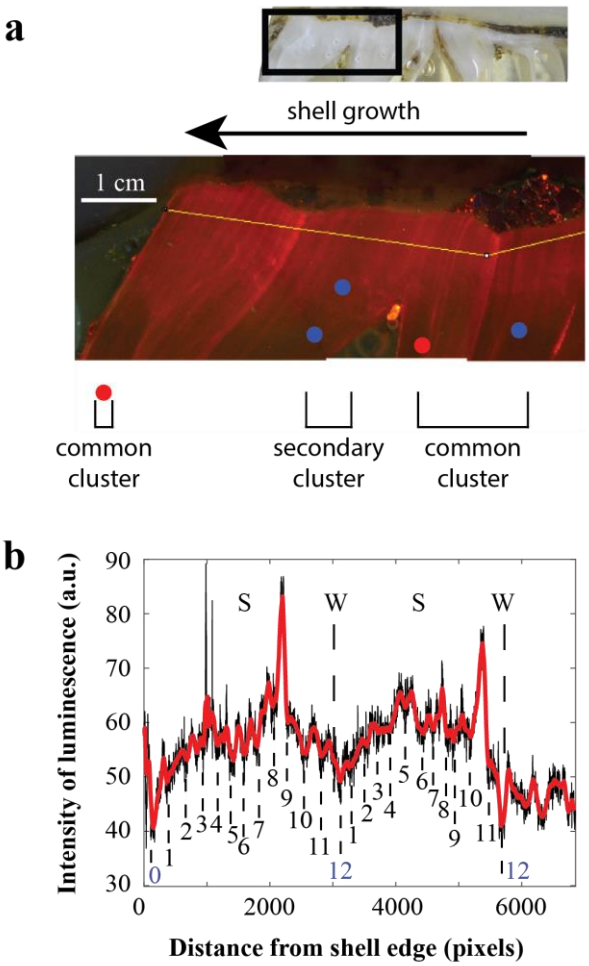

Figure 5: Successive measurements along shell growth indicating different fingerprints on specimen 1297-2 from Broue tower (Oe_BrT_A), namely "common cluster" and "secondary cluster" on Fig. 4b. **a:** Cathodoluminescence view of the last two years of growth (corresponding to the area analysed by LA-ICP-MS) on the umbo region and positions of the measurements. Red and blue dots correspond to summer and winter measurements, respectively. Note that the first measurement on the left is out of the picture frame. **b:** Intensity of luminescence (in arbitrary units) along the umbo following the line indicated on **a**, showing the areas of light (corresponding to summers; "S") and dull (corresponding to winters; "W") luminescence. Smoothing of this signal is shown overlapping in red, and highlights the presence of a monthly tidal cycle (numbers).

When considering the two remaining groups of confirmed origin, both from the Mediterranean Sea (an enclosed basin with a narrow opening to the Atlantic Ocean through the Strait of Gibraltar), each group is defined by a single cluster without any overlap (in green and orange in **Figure 4d**). The measurements from the groups of unknown origin overlap at least partially with the Oe_Mal_A group.

Data collected from specimens bred at Baie des Veys and Marennes-Oléron were used to study the differences between seasons for both species based on dendrograms (**Supplementary Information 5**). Seasons cannot be reliably discriminated, as indicated by the cluster analyses that show both seasonal measurements mixed together. For Cg_BdV_M, the cophenetic correlation coefficients are 0.80 and 0.52 for average and Ward methods, respectively. In the same site, for Oe_BdV_M, coefficients are 0.89 and 0.77 for average and Ward methods, respectively. Similar cophenetic correlation coefficients are found for Cg_MO_M with 0.70 and 0.62 for average and Ward methods, respectively, and for Oe_MO_M with 0.89 and 0.77 for average and Ward methods, respectively. The identification of seasons for all the measurements (except Leucate specimens; see section 2.2) in the t-SNE distribution is also reported in **Supplementary Information 5**.

4. DISCUSSION

4.1. Large-scale provenance discrimination

It was previously reported that REE and Y cannot be used for *O. edulis* as a provenance proxy due to vital effects, unlike *C. gigas* (Mouchi et al., 2020a). Consequently, other elements were investigated in this study in order to provide a potential regional fingerprint for this species and to find a way to efficiently compare measurements from *O. edulis* and *C. gigas* in terms of sourcing. The results show that REE are not the only elements whose incorporation differ between these two species. Indeed, the Cg_BdV_M and Oe_BdV_M groups, which originate from the same modern locality (and reared simultaneously on the same culture table in Baie des Veys, Normandy), exhibit obvious different compositions (**Figure 3**). Observations are similar for Cg_MO_M and Oe_MO_M from Marennes-Oléron, Charente-Maritime. However, it seems possible, when considering only one of the two species, to discriminate the composition depending on their region of origin, if not the locality.

For *C. gigas* specimens, the shells from the Mediterranean Sea (Cg_Leu_M and Cg_Dia_M), seem to be separated by t-SNE from the Atlantic Ocean/English Channel groups along the second dimension (approximately around unit 25 in **Figure 3**). This separation appears to be controlled by a

lower abundance in light REE in the Mediterranean groups compared to the Atlantic groups (**Supplementary Information 4**). This low abundance in light REE can either be specific to the Mediterranean shells or to the relatively small watersheds from these two localities (the smallest of the entire dataset). Although most of the Baie des Veys measurements are discriminated by t-SNE from other groups, some measurements share their composition with a part of the Tès group (Cg_Tes_M), which itself is partially connected to the Marennes-Oléron group (Cg_MO_M). This observation tends to indicate that specimens from the English Channel (Baie des Veys) and the Atlantic Ocean (Marennes-Oléron and Tès) exhibit some similarities in their geochemical composition.

For *O. edulis*, the common cluster (dashed ellipse in **Figure 4b**) gathers measurements from specimens from the Atlantic Ocean and English Channel coastlines, covering roughly 700 km of coastline (**Figure 1**) from Charente-Maritime in the south (Oe_MO_M, Oe_Bro_A, Oe_BrT_A) to Normandy in the north (Oe_BdV_M). As the English Channel and the Atlantic Ocean are connected across a wide area, the overall composition of seawater should share some similarities. On the contrary, the Mediterranean Sea is a semi-enclosed basin and is recharged by the Atlantic Ocean through the narrow Strait of Gibraltar. A difference in composition is expected in shells from Mediterranean localities. In addition, the near absence of tidal range in this region compared to the Atlantic Ocean can cause differences in fluid mixing and elemental incorporation in the shells. Therefore, it appears possible to discriminate, at a large scale, between specimens from the Atlantic Ocean/English Channel and the Mediterranean Sea.

4.2. Fine-scale provenance discrimination

The common cluster composed of samples from Oe_BdV_M, Oe_RsB_M, Oe_MO_M, Oe_Bro_A, and Oe_BrT_A shells (Baie des Veys: Normandy; Riec-sur-Belon: Brittany; Marennes-Oléron, Brouage and Broue tower: Charente) represents a homogenous fingerprint. These groups are from watersheds with quite distinct geological formations (limestones, basalts, etc…) but share a large seawater body (*i.e.*, the Atlantic Ocean). However, we observe that each group in this cluster has also a secondary cluster outside the common one, which seems group-specific, except for the Baie des Veys (Oe_BdV_M; **Figure 4b**). Within these groups, some specimens have all their measurements

within a single cluster (either the common cluster or the secondary cluster of the corresponding group), while other specimens have some measurements in both clusters (**Figure 5**). As these specimens have not been transferred from one location to another during growth, it appears that these changes in elemental incorporation occurred over the growth of the organism in the same site. As these changes occur multiple times over the growth period (**Figure 5**), it cannot be caused by ontogeny. This cyclical pattern is not linked to seasonality (this was checked by the seasonal calibration using cathodoluminescence; see t-SNE figure in **Supplementary Information 5**). Consequently, a different factor is at play. Temporal resolution of the LA-ICP-MS measurements is approximately one month, based on a tidal cathodoluminescence signal (Mouchi et al., 2013; see monthly cycles on **Figure 5b**) and *in vivo* labelling (Lartaud et al., 2010b; **Supplementary Information 6**). Growth rates can however be different according to the specimen and the period considered, and some measurements may represent a slightly shorter or longer period (Huyghe et al., 2019). Since all the corresponding groups originate from the English Channel and the Atlantic Ocean shorelines, tidal influence can be a pertinent factor. Indeed, all the groups from the Mediterranean Sea (Oe_Mal_A, Oe_Ale_A), which exhibits a very weak tidal range (only a few centimetres), are excluded from the common cluster (**Figure 4d**). In localities with a broad tidal range, the influence of the chemistry of nearby rivers could fluctuate widely, increasing during low tide and, on the contrary, being diluted with seawater at high tide. If this were the case, the secondary clusters would represent the correct local fingerprint, while the common cluster would correspond to the fingerprint of Atlantic seawater. Manganese is known to fluctuate in oyster shells from ebb to flood currents (Huyghe et al., 2019). Other chemical elements (*e.g.* those measured here) could present similar fluctuations. Switching between these two compositions, which are responsible for the presence of two distinct fingerprints, could be caused by the relative proportion (by volume in the analysed shell) of carbonate formed during ebb and flow currents. The proportion may vary, depending on growth rates. The case of the Baie des Veys shells (Oe_BdV_M group) is singular as it has no secondary cluster (**Figure 4b**). Interestingly, this site is also the locality with the most open marine settings from all modern groups. Although some shells of other groups did not have measurements corresponding to secondary clusters, the fact that no measurements from the 38 performed on five specimens are outside this common

cluster would tend to imply that there is only one fingerprint in this site. This observation is supported by the marine water mass current of the Baie des Veys, which flows towards the direction of the rivers' outputs from the locality where the oysters were collected (**Figure 1**; Lazure and Desmare, 2012), hence limiting the influence of the composition of the freshwater coming from the land. Becker et al. (2005) measured the elemental fingerprint of young mussel shells in several localities in San Diego County to study the larval dispersal along the coastline. They were able to observe distinct fingerprints for specimens from estuarine locations, but could not clearly discriminate open marine locations. We suspect this is what is observed here, with a 'marine' fingerprint common to several groups (being identical to that of an open marine locality, *i.e.* Baie des Veys), and a secondary fingerprint, specific to the locality, in environments where continental inputs are sufficiently strong. This assumption is supported by a higher and more stable salinity in Baie des Veys (approximately 33 all year) than in Marennes-Oléron (27 to 34; Lartaud et al., 2010c).

Temporal changes in the local elemental fingerprint have already been reported in mollusc shells, such as *Cerastoderma edule* and *Mytilus edulis* from Ireland (Ricardo et al., 2017; Bennion et al., 2019). These differences occur within months and observations in this study confirm that it is also the case for *O. edulis*, although the same is not true for *C. gigas*. These differences appear to switch back and forth between two fingerprints (**Figure 5**), one being common to multiple groups of shells of various origins in the French Atlantic Ocean and English Channel coastlines.

The case of the Charente-Maritime groups (Marennes-Oléron: Oe_MO_M; Brouage: Oe_Bro_A; and Broue tower: Oe_BrT_A) is interesting, as two distinct secondary clusters are visible in the t-SNE plot (**Figure 4c**). One of these groups of shells has been collected in the present day (Marennes-Oléron) with the exact location known, while the two other groups (Brouage and Broue tower) have been found in archaeological sites on land, with evidence that these locations were on the shoreline at the corresponding ages of the specimens. The fingerprint of the modern group is shared by the Brouage group dated from the 16th century CE (Oe_Bro_A). On the contrary, the Broue tower shells from the 11th and the 14th centuries CE (Oe_BrT_A) present a distinct fingerprint. It may be possible that the oysters from this group were living in a locality in the region under the influence of

another freshwater flow with a different water composition. Indeed, the Marennes-Oléron bay has two major tributary rivers, which cross different geological formations, although the main rock type is limestone (Bourgueil et al., 1968, 1972; Platel et al., 1977, 1978; Bambier et al., 1982; Hanztpergue et al., 1984; Mourier et al., 1989). The site of Brouage, from which were unearthed the shells of the Oe_Bro_A group, is located at the same level in the bay as the modern Marennes-Oléron group (Oe_MO_M; Champagne et al. 2012; **Figure 1**), with which it shares its secondary cluster. The site of the Broue tower, where the shells from the Oe_BrT_A group were discovered, is located further south, near the current output of the Seudre river (**Figure 1;** Normand et al., 2019). As seawater currents cross the bay from north to south, the oysters from the Oe_MO_M and Oe_Bro_A groups must have been under the influence of the Charente River (north side of the bay), but not the Seudre River. On the contrary, specimens from the Oe_BrT_A group must have been mainly under the influence of the Seudre River. The measurements show that, in such contexts, the identification of the region can be narrowed down to the watershed level. In addition to this, although temporal differences in local elemental fingerprints have been reported in other regions (Ricardo et al., 2017; Bennion et al., 2019), the fact that both modern (Oe_MO_M) and ancient (Renaissance, Oe_Bro_A) shells share a common cluster and a local cluster over the course of several centuries indicate that multiple measurements over the growth direction of shells can allow the local elemental fingerprint to be recognized, whatever the period of collection. This interpretation is also supported by the Oe_BrT_A specimens, which present fingerprints corresponding to both the common cluster and the same (locality-specific) secondary cluster, whether specimens are from the 11th or the 14th c. CE.

The only group of shells from the English Channel/Atlantic Ocean coastline that have no measurements corresponding to the common cluster is Oe_Lan_A. The Medieval site at Landévennec, from which these shells were unearthed, is located in the direct proximity of the Aulne River (**Figure 1**). Although the salinity is close to marine (generally around 32 except during flooding events where salinity drops to 15; Monbet and Bassoulet, 1989), the river rate of flow (annual mean of 24 $m^3.s^{-1}$; Auffret, 1981) is probably high enough to induce a strong influence in terms of chemical composition of the local seawater. That way, even at high tide, the composition of shells, derived from the local

seawater, would still be substantially influenced from the Aulne River. The two sub-groups from this cluster, with each corresponding to one specimen of different age (8th and 15th centuries CE), are probably due to a different degree of the influence of freshwater input. As several centuries separate these specimens, it is probable that the local geomorphology and water level changed, causing relocation of the supply of oysters from the inhabitants. Nevertheless, the close similar fingerprint of these specimens implies that they originate from nearby locations.

For *C. gigas* groups, all measurements from the Cg_Leu_M group of shells are clustered away from the other groups in **Figure 3**, with highly negative values on the 1st dimension axis and highly positive values on the 2nd dimension axis. This observation indicates that the elemental compositions of these shells are substantially different than the shells from the other regions. The fingerprint from the Diana lagoon (Cg_Dia_M) is also easily discriminated from the others. Compared to the other regions, these localities are characterized by small watersheds, uniquely consisting in limestone. As one can expect a smaller surface exposed to weathering than those of the other studied localities, the abundance of some elements in the shell should be lower at Leucate and Diana lagoon. Indeed, both groups are characterized by low abundances of light REE compared to the other groups, but the other analysed elements do not present such a systematic lower content (**Supplementary Information 4**). It is also possible that, as both these localities are in restricted lagoons, salinity can be at play. Indeed, Diana Lagoon (Mediterranean Sea) is known to have a high salinity (37.3 annual mean; Orsini et al., 2001) and Leucate lagoon has a seasonally dominated salinity (26 in winter and 42 in summer due to evaporation; Andrisoa et al., 2019), whereas Baie des Veys and Marennes-Oléron (Atlantic coastline) have lower salinities (33.4 and 32.4 annual mean, respectively; Lartaud et al., 2010c).

For "Atlantic Ocean" *C. gigas* specimens, the Baie des Veys group (Cg_BdV_M; although situated in the English Channel, with a wide access to the Ocean) appears to be the most distinct by t-SNE between these groups, with only some measurements overlapping with the Tès group. This is interesting, as *O. edulis* shells from the same locality (Oe_BdV_M) had the peculiar characteristic of representing the composition of a cluster common to other Atlantic groups. The Tès (Cg_Tes_M) and Marennes-Oléron (Cg_MO_M) groups share their distribution in the t-SNE graph, with the Marennes-

Oléron distribution entirely comprised into a restricted portion of the Tès distribution. The large distribution by t-SNE of Tès measurements compared to Marennes-Oléron can be explained by the annual range of salinity (approx. 15 and 5, respectively for Tès and Marennes-Oléron; Lartaud et al., 2010c). These two localities (Arcachon Basin and Charente-Maritime) have their watersheds crossing different geological formations with mainly Cenozoic river deposits for Tès (Dubreuilh et al., 1992) and mainly Mesozoic limestone for Marennes-Oléron (Bourgueil and Moreau, 1967, 1970; Bourgueil et al., 1968; Platel et al., 1977; 1978). Still, the deposit cover of the respective watersheds does not appear to be sufficient to entirely discriminate these groups.

4.3. Attempting to identify the provenance of the Lyons/Cybèle shells

The Oe_Mal_A group from La Malène belongs to a specific locality, which is unknown, but at least confirmed from the Mediterranean coastline, probably from Languedoc coastline due to the associated remains from other taxa specific to Roman-influenced Languedoc customs (Bardot-Cambot and Forest, 2014; Forest, 2019; Forest, 2020 for Mureau, 2020). Measurements from this group are discriminated from the other *O. edulis* groups of known origins (all from the Atlantic Ocean/English Channel coastlines) and these groups are relatively close to one another. We note that the Oe_X1_A and Oe_X2_A group distributions are partially overlapping (**Figure 4d**). It seems that the shells from these groups of unknown origin, and found at the Antiquity Cybèle site (Lyons, over 200 km away from the nearest shore), share part of their fingerprint, which could imply that all these specimens lived in the same geographic region, yet not the exact same locality. Therefore, although geochemistry confirms that the Cybèle groups initially determined by morphometric parameters (Bardot-Cambot, 2013) do exist, our data tend to discard the hypothesis that one group originated from the Mediterranean Sea and the other from the Atlantic Ocean. Although additional sites from both Atlantic and Mediterranean shorelines would be required to affirm this, it seems here that populations from these groups lived a short distance away from each other, and the fingerprint of the Oe_X1_A group seems to correspond to that of La Malène specimens (Oe_Mal_A). If true, additional specimens in other localities on the Mediterranean shoreline should be analysed to identify their locality of origin, but at the moment, we were not able to collect wild modern specimens.

4.4. Quality of the discrimination method

As our dataset is constituted of 14 elements per measurement, attempts were made to reduce this number for ease of analytical and data processing work. Some elements were found to be of increased importance compared to others to adequately separate measurements from different groups using t-SNE. By successive removal of one element from the dataset, Pb, Ba and U are the most effective at successfully discriminating *O. edulis* groups from the Mediterranean Sea and the Atlantic Ocean (**Supplementary Information 7**). Rare Earth Elements (La, Ce, Pr, Nd, Sm), as found by Mouchi et al. (2020a), were most useful for discriminating *C. gigas* groups. All other elements were found to contribute equally to discriminating specific localities. In the end, the removal of any of the measured elements systematically reduces the quality of the discriminating method.

Cross-validation of the clusters was attempted on *O. edulis* measurements (without the groups of unknown origin, *i.e.* Oe_X1_A and Oe_X2_A) using discriminant analysis. We obtained a cross-validated classification error of 24.3% (out of 210 measurements). However, when we removed all measurements from the common cluster (putatively corresponding to the open marine fingerprint, including all measurements from the Oe_BdV_M group) from the comparative dataset, we obtained a cross-validated classification error of 0.8% (out of 123 measurements from 7 groups). Becker et al. (2005) were able to retrieve an open marine origin from San Diego County mussel shell composition with a 5% error (out of 41 measurements), which corresponded to misclassification to open bays. However, although these specimens had been collected from six open marine localities along the Pacific Ocean coastline of San Diego, the correct identification of the site had a 44 % error. Misclassification of specimens from two open bays (between both bays and all open marine localities as a whole) presented errors of 33 and 62 % (out of 6 and 8 measurements, respectively) due to a misclassification of measurements to open marine environments. These results indicated that the open marine fingerprint from the mussel shell composition is similar along 50 km of coastline (range of the collection sites of the study), and that elemental fingerprinting of specimens from intertidal localities in bays present some similarities to the regional open marine fingerprint. Our observations concur with these conclusions, albeit with additional precision.

5. CONCLUSION

Previous attempts to discover the geographical origin of archaeological oyster shells based on geochemistry presented various degrees of success due to the use of isotopic ratios that are strongly influenced by local environmental conditions, which can be similar over extremely remote areas. In this study, elemental fingerprinting of oyster shells from 15 different groups was used, highlighting several points. First, different elemental incorporation behaviours prevent the direct comparison of different species. Second, cluster analysis from the t-SNE method suggests that a regional fingerprint seems to exist and to be distinguishable from large water bodies (seas and oceans). Third, in environments with restricted influence of oceanic water, a more precise fingerprint is identified and the locality (at the watershed scale) can be found. In addition, as previous studies reported, none of the elements is able to differentiate between the origins of various groups on its own. Last but not least, this technique was successful to identify the provenance of oyster shells of known and previously unknown origin from archaeological sites. Therefore, this technique appears adequate and promising to pinpoint the provenance of oyster shells. It remains necessary, however, to have the elemental fingerprint of a multitude of coastal sites as a database, so that the fingerprint of unknown specimens can be recognized.

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511

512 7. Data availability

513 The data used in this study have been deposited in the Zenodo data repository
514 (http://doi.org/10.5281/zenodo.4681223; Mouchi et al., 2021).

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516 Declarations of interest: none.

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