Journal of Archaeological Science: Reports 17 (2018) 242–249

Contents lists available at ScienceDirect

Journal of Archaeological Science: Reports

journal homepage: www.elsevier.com/locate/jasrep

Geochemical markers of human occupation in the lower Argens valley (Fréjus, France): from protohistory to Roman times

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ARTICLE INFO

Keywords:
Geoarchaeology
Fréjus
Roman harbor
Pollutant metals
Lead isotopes
Iron Age

ABSTRACT

Sediments deposited in the lower Argens valley at Fréjus, a well-known Roman Imperial harbor in northwestern Mediterranean, reveal pollutant metal imprints since the Iron Age. Lead, copper and zinc concentrations in sediments above a facies transition from sand (lower) to silt (upper) at 1820–1946 cal. BP (65–130 cal. CE) are four times higher than in the non-contaminated geochemical background unit at the bottom of the core. These correlated chemical and sedimentological shifts in the ancient marine bay follow the growth of the Augustan city subsequent to the building of the Roman harbor during the late first century BCE. Trace metal enrichment factors (calculated from background concentrations) display a slight but significant increase (1.3 to 1.5) in sediments deposited during the Fréjus protohistoric period, just below the facies transition. This enrichment is corroborated by mean $^{206}Pb/^{207}Pb$ isotope ratios that vary from 1.179 (± 0.002) to 1.193 (± 0.002) and 1.202 (± 0.002) in the Roman, protohistoric and background units respectively. These findings are the first evidence of pollutant metal release into the lower Argens ria during the protohistoric period, as far as 2600 years ago. Lead isotope signatures in the protohistoric sediment layers shed light on several possible geographic origins for ores from which accumulated pollutant metals may originate. These include copper mines in the Alps or the Languedoc regions. Cyprus may also be isotopically invoked as a source that is not supported by archaeological findings and would constitute, if proven true, a new insight regarding metal trades in protohistoric Gaul.

1. Introduction

Fréjus is located in southeastern France, in the distal margin of the Permian basin of the Argens river (Fig. 1a), at the mouth of an alluvial Mediterranean delta of which extension evolved according to combined climatic and anthropogenic interactions during the Holocene (Bertoncello et al., 2014; Bertoncello and Devillers, 2006; Dubar, 2004). The 114 km long low-sloping Argens river has a watershed of 2500 km^2, with mostly non-navigable waters. The Romans took advantage of Fréjus geographic and geomorphologic assets with the proximity of the connecting road between Ligurian Italy and the Lower Rhône valley, the existence of an opened marine bay and a sandstone promontory that protects the city from riverine flooding (Bony et al., 2011; Chevallier, 1998; Devillers et al., 2007; Excoffon et al., 2006; Gébara and Morhange, 2010) to establish *Forum Iulii* during the first century BCE. (Gascou, 1982; Gascou and Janon, 1985). The foundation of *Forum Iulii* would not be older than 49 BCE, when a Forum was established by Jules Cesar (Brun, 1999; Duval, 1971; Gascou and Janon, 1985). Historical and epigraphical sources suggest that Fréjus acquired the status of Roman colony with Octavius around 30 BCE (Christol, 2015; Gascou, 1982) with the establishment of legion veterans, not long after the arrival of the captured Mark Antony fleet after the defeat of Actium in 31 BCE. The development of *Forum Iulii* is described by many archaeological studies (building remains, soundings, geophysical prospection, artifacts, excavations) (see references in Gébara, 2012; Pasqualini, 2011; Rivet et al., 2000).

While the Roman colony has left numerous archaeological imprints and has clearly modified the local geomorphology, there are much fewer indications of occupation at Fréjus itself during the Iron Age (between the VIIIth and the Ist century BCE.) (Bérato et al., 1991; Brun, 1999; Excoffon et al., 2010a; Rivet et al., 2000). Some protohistoric remains were found in the Saint-Antoine hillcock (portion of a wall associated to pre-roman pottery) (Rivet et al., 2000) but they are too scarce to identify an occupation of some importance at Fréjus before the

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http://dx.doi.org/10.1016/j.jasrep.2017.09.028
Received 16 June 2017; Received in revised form 5 September 2017; Accepted 30 September 2017
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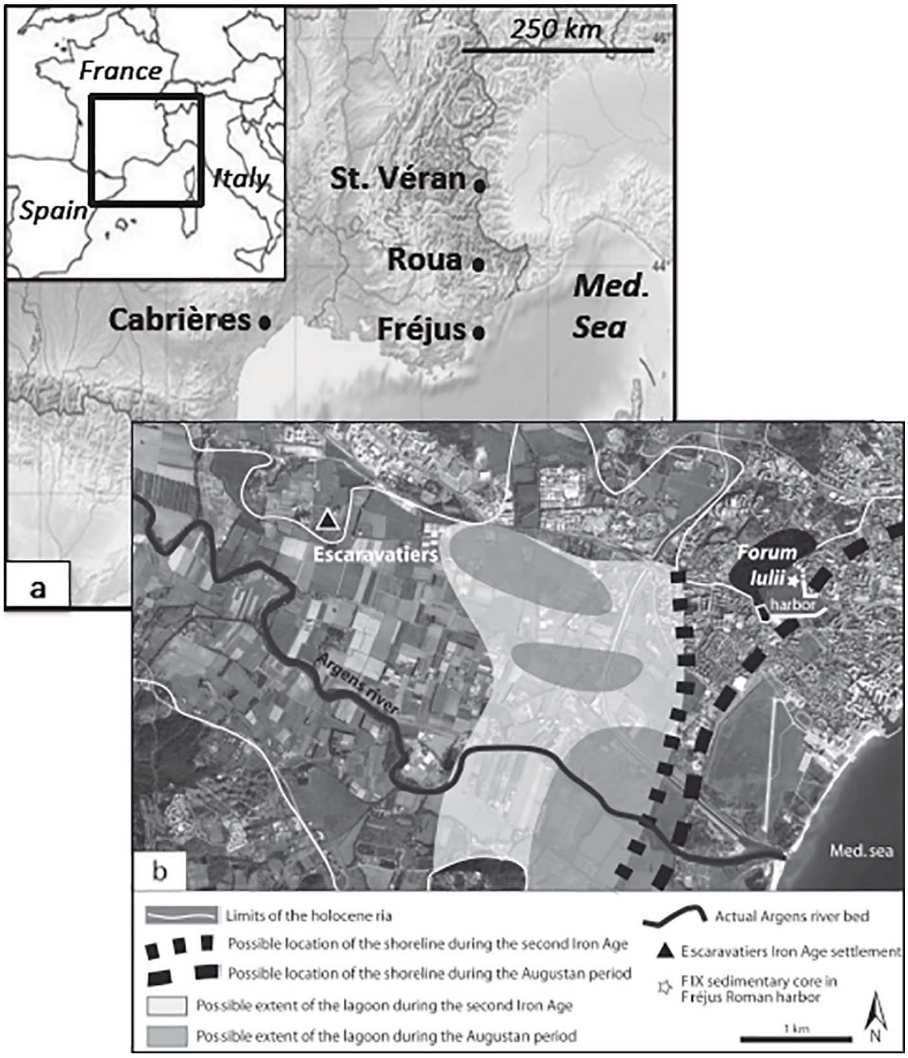

Fig. 1. a) Geographical location of Fréjus and Cu mining sites that are described in the text; b) Palaeogeographical reconstruction of the lower Argens valley during the second Iron Age and the Augustan period (from Bertoncello et al., 2014) with the location of FIX sedimentary core in Fréjus Roman harbor and of the Escaravatiers Iron Age settlement.

founding of the *Forum Iulii*. On the contrary Iron Age occupation is well evidenced outside of Fréjus in the Argens valley, about 10–30 km upstream from the delta (Bérato and Magnin, 1989; Bérato et al., 1990; Bérato et al., 1991; Bertoncello, 1999, 2005; Pellissier, 2006). Protohistoric habitats were identified within 15 km of Fréjus at the Arc-sur-Argens, Roquebrune-sur-Argens and Le Muy (Brun, 1999). The closest and most developed settlement (1,5 ha) is located at Puget-sur-Argens, at the Escaravatiers Highs, 3 km north of Fréjus (Fig. 1b). It was occupied from the end of the VIth c. BCE to likely the first quarter of the Ist c. BCE (Fiches et al., 1995). The quantity and quality of the archaeological remains recovered by field survey on the site and its continuous occupation for more than 4 centuries, suggest that it was one of the major settlements in this part of eastern Provence during the Iron Age, although its exact form and function remain unknown as no archaeological excavation was conducted on the site. At the time of its occupation, the site may have been only a few kilometers away from the seashore (Fig. 1b; Bertoncello et al., 2014; Devillers and Bonnet, 2006; Dubar, 2004; Fiches et al., 1995). Paleoenvironmental proxies also reveal protohistoric occupation along the Argens river in the lower valley, meanwhile, no archaeological evidences have been found in the ancient alluvial delta, possibly owing to erosion or sedimentary burying due to prograding processes (Bertoncello et al., 2008, 2014; Provansal et al., 2006).

Nowadays, the chronology of Fréjus foundation and the growth of the initial Forum and the Colony are acknowledged, meanwhile, the initial operating of the harbor remains uncertain. This is in part due to its layout in a opened marine bay as shown by paleoenvironmental results that include sedimentology, granulometry, palynology and fauna assemblages records among others (Bertoncello et al., 2014; Bony et al., 2011; Devillers et al., 2007; Excoffon et al., 2010a, 2010b; Excoffon et al., 2016). As such these investigations aim at defining the paleogeographical evolution of the shoreline in relation to land occupation, marine dynamics and riverine progradation since the Neolithic period and the landscape that existed at the foundation of the city during the first century BCE (Bertoncello et al., 2014; Bony et al., 2011; Devillers et al., 2007; Excoffon et al., 2006; Gébara and Morhange, 2010).

The foundation and development of Ancient cities is usually associated with the accumulation of pollutant metals into nearby soils and sediments owing to aerosol deposition and water run-off particle dispersion from forge/metallurgical activities as shown in several coastal lagoons and harbors during the Antiquity (Delile et al., 2014; Elmaleh et al., 2012; Le Roux et al., 2003, 2005; Stanley et al., 2007; Véron et al., 2006; Veron et al., 2013). Metals may not only be complementary markers of *Forum Iulii* growth but also of protohistoric human activities in the lower Argens valley. Indeed, protohistoric sepulchers and habitats have divulged a few metal artifacts, mostly bronze made, that included jewels, coins and weapons (Bérato et al., 1991; Brun, 1999; Excoffon et al., 2010a, 2010b; Fiches et al., 1995) suggesting the use of copper (Cu) ingots and/or local forges that may have left imprints in soils and sediments of the valley.

Here we propose to investigate pollutant metal accumulated within the Roman harbor of Fréjus as indicators of human activities to discern chemical imprints of the city growth and nearby protohistoric activities. While trace metals are found enriched in many Ancient harbors, mineralogical and sedimentological variations may partly conceal anthropogenic imprints, most particularly when metal enrichments are faint. In such case, ratios of stable lead (Pb) isotopes have been found efficient to evidence the use of metals and/or smelting technologies in Mediterranean harbors (Delile et al., 2014; Le Roux et al., 2003, 2005; Veron et al., 2013). There are 4 stable Pb isotopes, ^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb, the last three being end-members of the natural Uranium (U)-Thorium (Th) decay chains. As such, the relative amount of these isotopes varies in different Pb and Cu ores according to the initial Th and U concentrations and the age of the ore bodies (Doe, 1970). Crustal Pb deposits found in marine unconsolidated sediments originate from natural geological erosion and display specific isotopic imprints that are different from those of ores. Not only Pb isotope imprints allow to discriminating the crustal ("natural") and ore ("anthropogenic") origin of Pb encountered in sediments, but they also provide insights on the geographic origin of ore bodies from which accumulated metals have been mined (Brill and Wampler, 1967; Gale, 1999, 2001; Gale and Stos-Gale, 1982, 2000; Philip et al., 2003; Sayre et al., 2001; Spooner and Gale, 1982; Stos-Gale et al., 1997; Véron et al., 2012, 2013). Pb (and most other metal contaminants) is proven immobile once deposited and buried into marine sediments, as shown with the comparison of Pb and its corresponding stable isotope imprints in ^{210}Pb/^{14}C dated cores to atmospheric time series and/or specific deposition events, and therefore is an efficient marker of past anthropogenic activities as revealed from well-dated levels (Ferrand et al., 1999; Miralles et al., 2006; Véron et al., 2006; Angelidis et al., 2011). As such we expect to chemically characterize (1) the development of the Roman city and most particularly of its harbor and (2) the impact of protohistoric communities, owing to the release of metals and corresponding isotope markers into their nearby environment.

2. Methods

We have chosen to investigate chemical markers from a core collected within the ancient harbor of *Forum Iulii*, at the bottom of the Saint-Antoine hillcock promontory (Fig. 1b), as part of the ANR program Paleomed. This 9 m core (FIX) was sampled along with two other sedimentary cores by Bony et al. (2011) in order to answer two main paleoenvironmental questions, i.e. the genesis and evolution of the Roman harbor in relation to riverine and/marine progradation and how natural coastal constraints and hazards have impacted upon the harbor. Combining archaeological and geomorphological data the authors reconstructed paleogeographic and chrono-stratigraphic evolution of the marine bay (Bony et al., 2011). A detailed stratigraphic log of the core can be found in Bony et al. (2011). Briefly, FIX sediment core is characterized by two main lithologic units including coarse sands (9.1–6.8 m) and silts (6.8 m and above) that suggest different sediment deposition dynamics. The transition from sand to silt facies sediments at 6.8 m (1820–1946 cal. BP) also corresponds to a biostratigraphic shift from subtidal and benthic marine macrofauna to ostracod (*Cyprideis torosa*) dominated lagoonal biocenoses (Bony et al., 2011). This sudden change from a protected marine cove to an euryhaline open lagoon is seen within the three sediment cores collected in the harbor basin (Bony et al., 2011). This paleoenvironmental shift may be linked to the layout of the port infrastructures, although the chronological span given by the radiocarbon date is too wide to precisely date the building of the Roman harbor (120 cal BCE–60 cal CE, Bony et al., 2011). Following this transition, Bony et al. (2011) evidence several storm events, progradation of the Argens river and subsequent landlocking of the harbor that may have led to anthropogenic dredging of the basin, as suggested by the lack of sediment deposition record during the first centuries CE. Here we shall focus our chemical investigation on core FIX around (1) the transition from sand to silt facies at 1820–1946 cal. BP as identified by Bony et al. (2011) and (2) the sediment record at the bottom of the core in order to detect any anthropogenic imprint that may have predated the founding of *Forum Iulii*. A total of nine ^{14}C dates were measured at Poznan Radiocarbon laboratory in Poland. Age calibration was performed using Oxcal V3.10 with IntCal04 (for wood, grains, continental organic matter) and Marine04 (for marine shells, posidonia). ^{14}C age model for sediment deposits are presented in cal. BP. (calculated Before Present, i.e. before 1950, prior to the atmospheric dispersion of ^{14}C from nuclear tests). More details regarding the stratigraphy of core FIX can be found in Bony et al. (2011).

Sediments were subsampled from core FIX of which exact location in Fréjus harbor basin and sedimentological description can be found in Bony et al. (2011). About 50 mg of each sample were dissolved with concentrated acids (HCl, $HN0_3$ and HF) at 130 °C. A fraction of the digested sediment was diluted in 1% $HN0_3$ before Pb, Cu (copper), Zn (zinc) and Ti (titanium) analysis by Inductively Coupled Plasma Mass Spectrometry (ICPMS Perkin Elmer NexIon 300X) at CEREGE. Total blanks accounted for less than 1% (Pb, Ti) and 5% (Cu, Zn) of measured amounts. Analytical precision was below 3% for all measured elements. Metal concentrations are shown in Table 1 and Fig. 2 with corresponding depths and calibrated ages determined from ^{14}C dating (see Bony et al., 2011). Stable lead isotope ratios were determined by Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICPMS Isoprobe) at GEOTOP (University of Quebec at Montreal) on the other digested sediment fraction following AG1X8 resin extraction as described in Manhès et al. (1978). Isotopic ratios were normalized to multiple measurements of the NBS981 standard (Doucelance and Manhès, 2001). Long-term reproducibility assessment from internal standard measurements was better than 0.03% for all reported Pb

Table 1
Trace metal concentrations ($\mu g \cdot g^{-1}$ or ppm) and enrichment factors (EF) for sediment deposits of core FIX with depth (m) and corresponding ages (years cal. BP). Note that trace metal concentrations are missing for samples FIX28 and FIX46.

Sample. Ref.	Depth (m)	Age (cal. BP)	Ti (ppm)	Pb (ppm)	Cu (ppm)	Zn (ppm)	EF_{Ti} (Pb)	EF_{Ti} (Cu)	EF_{Ti} (Zn)
FIX12	3.1	181	2496	91.5	28.6	109	3.0	3.6	2.4
FIX21	4.5	754	2205	94.6	44.7	118	3.5	6.2	2.9
FIX32	6.3	1879	2426	102	23.9	92.0	3.4	3.0	2.0
FIX33	6.4	1857	1858	65.9	14.7	45.0	2.9	2.4	1.3
FIX36	6.8	2027	2101	35.1	8.74	40.0	1.4	1.3	1.0
FIX39	7.1	2162	1849	24.7	8.25	44.9	1.1	1.4	1.3
FIX40	7.2	2207	2085	32.9	10.0	48.1	1.3	1.5	1.2
FIX43	7.6	2342	1013	17.1	6.48	40.0	1.3	1.9	2.1
FIX47	8.4	2628	1479	15.9	3.95	25.5	0.9	0.8	0.9
FIX49	8.6	2718	1364	19.3	5.64	30.5	1.1	1.3	1.2
FIX52	8.9	2815	1814	18.6	5.30	30.8	0.8	0.9	0.9
FIX54	9.1	2907	772	12.8	2.57	13.2	1.3	1.0	0.9

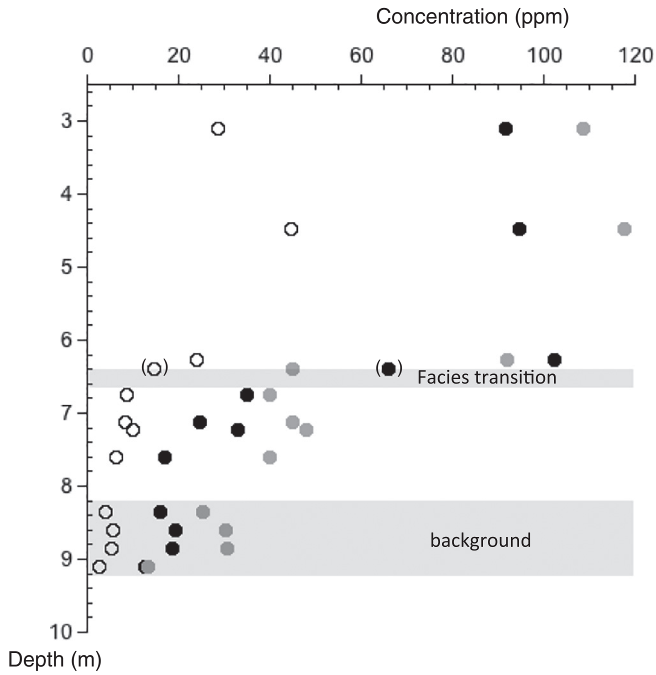

Fig. 2. Trace metal concentrations (Cu-open circles; Pb-dark closed circles; Zn-grey closed circles) with depth (m) in core FIX. The sand to silt "facies transition" grey unit is defined in core FIX from Bony and al. (2011) paleoenvironmental study. The "background" grey unit is defined from trace metal concentrations measured in the core as part of this study.

isotopic ratios. Because of their precision and sensitivity $^{206}Pb/^{207}Pb$ ratios have been very much used in the literature and shall be presented in our discussion. Taking into account the analytical precision and the observed natural variation in $^{206}Pb/^{207}Pb$ isotope ratios, differences above 0.2% (an order of magnitude above the analytical precision) may be considered geochemically significant to discuss isotope-based shifts in anthropogenic and/or crustal sources. Ratios of stable Pb isotopes are shown in Table 2 and Fig. 3. Ti was used as crustal reference to calculate Enrichment Factors (EF) for each metal (Me) as the ratio of Me/Ti in each sample to that of a defined "natural" background in the core (see De Vleeschouwer et al., 2007, 2009; Shotyk et al., 2002 for more details regarding the use of Ti as crustal reference). This background is defined with the less likely contaminated samples for concentrations and stable lead isotope ratios. The degree of contamination relies on EF values: with EF close to 1, there is no enrichment versus the crustal contribution, with EF < 3, a minor enrichment may be considered, while EF > 5 are indicative of a significant non-crustal input of exogenous metal, likely due to anthropogenic activities (see Py et al., 2014; Veron et al., 2013 for archaeological relevant application). EF for Pb, Cu and Zn are presented in Table 1 and Fig. 3.

3. Metal enrichments in sediment deposits

A well-defined decreasing trend is evidenced for trace metal content (Tab. 1, Fig. 2) with concentration ranging between 95–16 ppm, 45–2.6 ppm and 118–25 ppm for Pb, Cu and Zn respectively. These anthropogenic markers display a sharp significant decrease at 6.2–6.4 m into the core (student *t*-test $p \leq 0.05$) with mean concentrations varying from Pb = 87.6 ± 19.3 ppm, Cu = 32.4 ± 10.9 ppm, Zn = 106 ± 13 ppm at the surface (intermediate bracketed values in Fig. 2 are not taken into account for Pb and Cu mean calculations) to Pb = 22.8 ± 8.5 ppm, Cu = 6.4 ± 2.5 ppm, Zn = 35.3 ± 11.3 ppm below the interface between marine sands and silt deposit facies (Fig. 2) as identified by sedimentological and fauna assemblage at 6.4–6.6 m depth in core FIX (Bony et al., 2011). This concentration change at 1820–1946 cal. BP may result from the building of the Roman harbor

Table 2
Pb isotopic ratios in core FIX with depth (m) and corresponding ages (years cal. BP).

Sample. Ref.	Depth m	Age cal. BP	$^{206}Pb/^{207}Pb$	$^{208}Pb/^{206}Pb$	$^{206}Pb/^{204}Pb$
FIX12	3.1	181	1.1788	2.0880	18.462
FIX21	4.5	754	1.1781	2.0907	18.450
FIX28	5.8	1323	1.1768	2.0936	18.416
FIX32	6.3	1879	1.1832	2.0850	18.538
FIX33	6.4	1857	1.1781	2.0886	18.440
FIX36	6.8	2027	1.1922	2.0832	18.682
FIX39	7.1	2162	1.1937	2.0788	18.702
FIX40	7.2	2207	1.1941	2.0786	18.732
FIX43	7.6	2342	1.1953	2.0745	18.742
FIX46	8.2	2583	1.1911	2.0795	18.661
FIX47	8.4	2628	1.2049	2.0764	18.896
FIX49	8.6	2718	1.2007	2.0754	18.838
FIX52	8.9	2815	1.2012	2.0776	18.841
FIX54	9.1	2907	1.1997	2.0813	18.828

and the growth of the Augustan city. This result is consistent with local paleoenvironmental reconstructions (Bony et al., 2011) and proposed dating of the oldest harbor infrastructures (Gébara and Morhange, 2010). There is a more subtle trend below this transition (student t test $p \leq 0.05$), at 8 m, with mean concentrations shifting from Pb = 30.9 ± 5.5 ppm, Cu = 8.4 ± 1.5 ppm, Zn = 43.6 ± 3.5 ppm above 8 m to Pb = 16.7 ± 3.0 ppm, Cu = 4.4 ± 1.4 ppm, Zn = 35.0 ± 8.2 ppm below (Fig. 2). The deepest least concentrated fraction (below 8 m) may be considered as the geochemical background of the core with little or no contaminated particles (Fig. 2). The mean background value calculated for Pb (16.7 ± 3.0 ppm), a highly sensitive and well-monitored anthropogenic marker, is within the expected crustal concentrations measured in non-contaminated sediments from coastal Western Mediterranean areas (13–20 ppm; Ferrand et al., 1999; Miralles et al., 2006; Roussiez et al., 2005).

The use of trace metal concentrations alone may be ambiguous because of natural variations resulting from changes in the mineralogical content of the sediment core. This uncertainty can be partly overcome with Enrichment Factors (EF) normalization and the use of a naturally occurring element (Ti) as described in the "Methods" section. We calculate EFs for Pb, Cu and Zn from mean concentrations measured in the background section below 8 m as defined previously (Figs. 2 and 3) with Ti = 1354 ± 428 ppm, Pb = 16.7 ± 3.0 ppm, Cu = 4.4 ± 1.4 ppm and Zn = 35.0 ± 8.2 ppm. Taking into account the standard deviation of these mean concentrations, we consider no contamination enrichment in sediment deposits with EF below 1.2 (Fig. 3). EFs are consistent with concentration trends, and are significantly (student *t*-test $p < 0.05$) above unity in the silt facies unit, down to the transition area at 1820–1946 cal. BP. (Fig. 3). Mean EFs shift from 3.2 ± 0.3 (Pb), 3.8 ± 2.8 (Cu) and 2.1 ± 0.7 (Zn) in the top silt facies unit to 1.3 ± 0.1 (Pb), 1.5 ± 0.1 (Cu) and 1.4 ± 0.4 (Zn) in the sand facies unit, just above the background. It should be noticed that EF differences are statistically significant for Pb and Cu (student *t*-test $p \leq 0.02$) between the silt and the sand facies, but not for Zn ($p = 0.1$) owing to substantial standard deviations for mean concentrations of the latest. Trace metal concentrations and corresponding EFs not only suggest a significant anthropogenic imprint starting at 1820–1946 cal. BP in Fréjus, supposedly related to the development of the Roman harbor and the Augustan city, but also suggest a protohistoric imprint between 2000 and 2350 years cal. BP (Table 1, Figs. 2 and 3). Using concentrations one may calculate the anthropogenic fraction (Me_a) for each enriched metal as followed:

$$Me_a = [Me_s - Ti_s(Me/Ti)_b]$$

With Me_s and Ti_s being the amount of Me and Ti in the sample, while $(Me/Ti)_b$ is the ratio of Me to Ti in the background unit. The excess anthropogenic fraction in the well-established Roman harbor is 40–70 ppm (Pb) and 9–16 ppm (Cu) while it reaches 2–9 ppm (Pb) and

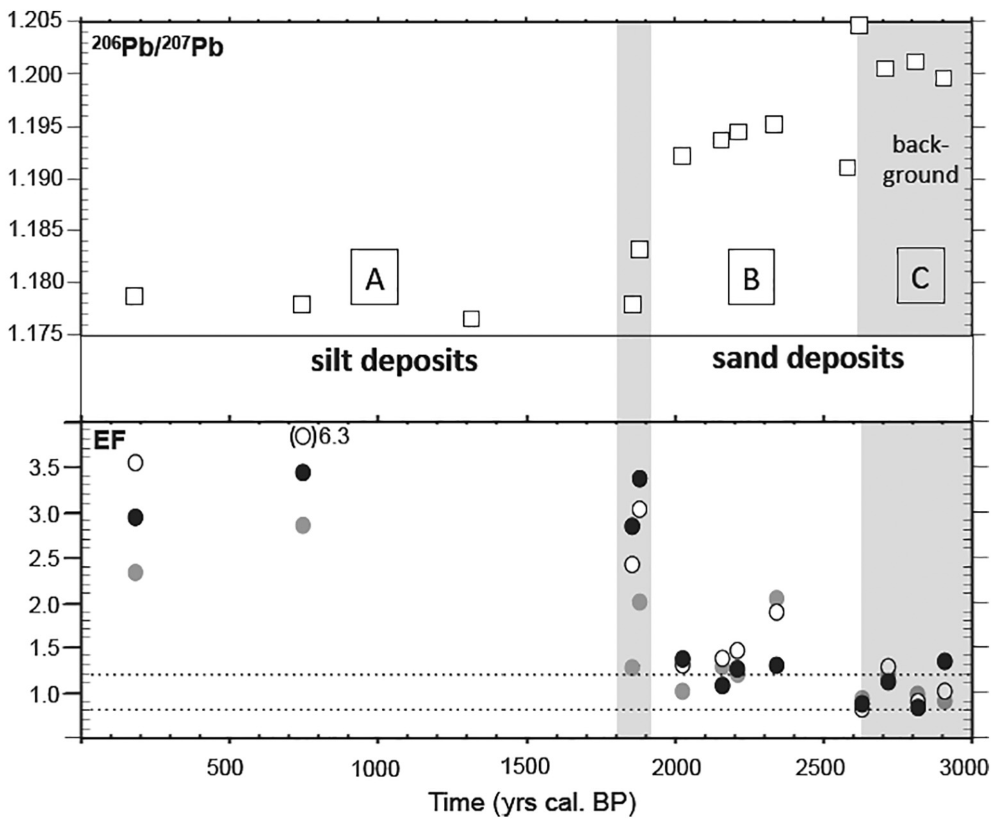

Fig. 3. Trace metal enrichment factors (EF) and $^{206}Pb/^{207}Pb$ ratios are shown with time (years cal. BP) as defined from ^{14}C age model in core FIX (Bony et al., 2011). The background and transition grey band units are similar to those shown in figure 2. Caption for Cu, Pb and Cu are the same as those used in Fig. 2. Sections A, B, C are shown as defined in the text. Note that one EF for Cu is shown in parenthesis with its value (6.3) in order to keep the figure at an appropriate scale for all other EF values.

2–3 ppm (Cu) in the protohistoric sediment layers. Pollutant metal accumulation in the Roman harbor largely exceeds that during the preceding Iron Age as expected from previous findings in Mediterranean Ancient harbors (Le Roux et al., 2003, 2005; Véron et al., 2006). Meanwhile, Pb enhancement between the two periods (5–35 times) is much larger than that for Cu (3–8 times) suggesting a largest paleoenvironmental contamination than expected during the protohistoric period for Cu.

The choice of a given crustal reference may affect EF values depending on the mineralogical content of the core (Huisman et al., 1997). Here Ti may be influenced by Ti rich minerals such as rutile, ilmenite or titanite, particularly in fined silt sediments as those encountered in the contaminated top core unit above the 1820–1946 cal. BP transition. Such Ti enrichment would lower EF calculated in this top unit, and therefore our EF estimates would then be minimal, suggesting an even larger contamination impact from the Roman city, and therefore greater enrichments versus the protohistoric levels. In order to confirm and investigate more thoroughly chemical imprints during the Iron Age, we have analyzed other chemical markers, stable Pb isotope ratios, that are very efficient to evidence metal-associated anthropogenic activities as explained in the “Introduction” section.

4. Isotopic evidence of protohistoric imprints

Lower concentrations at the bottom of FIX core, within the geochemical background unit, are consistent with more radiogenic Pb isotopic imprints (mean $^{206}Pb/^{207}Pb = 1.202 \pm 0.002$ at 8.4–9.1 m, FIX47 to 54) (Table 2, Fig. 3) that are similar to those from non-contaminated sediments in the Western Mediterranean (Angelidis et al., 2011; Ferrand et al., 1999; Miralles et al., 2006). Isotopic ratios confirm the choice of the geochemical background based on concentration alone. Cu and Pb ores (and artifacts made of) generally produce less radiogenic isotope signatures, i.e. lower $^{206}Pb/^{207}Pb$ ratios (see ref. in Veron et al., 2013). The $^{206}Pb/^{207}Pb$ ratios display a significant trend in the core (student *t*-test $p \leq 0.002$) from (A) 3.1–6.4 m ($t < 1850$ years cal. BP) (mean $^{206}Pb/^{207}Pb = 1.179 \pm 0.002$), to (B) 6.8-8.2m ($2027 < t < 2583$ years cal. BP) (mean $^{206}Pb/^{207}Pb = 1.193 \pm 0.002$) and (C) 8.4–9.1 m ($t > 2628$ year cal. BP) (mean $^{206}Pb/^{207}Pb = 1.202 \pm 0.002$). These three geochemical units are consistent with the sedimentological-fauna based facies units observed by Bony et al. (2011) and trace metal concentrations measured in core FIX (Fig. 3). Unit (A) closely fits within the well-identified silt unit while (B) and (C) correspond to sub-units we have identified in the marine sand facies, i.e. the protohistoric imprint and the geochemical non-contaminated background. Mean $^{206}Pb/^{207}Pb$ ratios confirm the occurrence of a protohistoric imprint in Fréjus ancient bay, between approximately 2000 and 2600 year cal. BP. From these results we may infer that protohistoric occupation contributed to the release of pollutant metals into the lower Argens delta during the second Iron Age, prior to the founding of *Forum Iulii*. These significant geochemical imprints likely result from the use of metal ingots and forges rather than ore treatment since no Cu or Pb mines and metallurgical relics have been found within 50 km of Fréjus (Brun, 1999; Pagès, 2008). The dispersion of such contamination into Fréjus marine bay could result either from local aerial dispersion and/or run-off from protohistoric settlements located on highs or along opened coastal lagoons nearby the coastline during the Iron Age (Fig. 1b). Additional archaeological and paleoenvironmental investigation are needed to ascertain the role of the Escaravatiers site and/or reveal further Human occupations within the lower valley during the protohistoric period.

The remarkable correlation between Cu, Zn and Pb concentration in sediments suggest a polymetallic ore source of which geographic origin may be investigated thanks to the use of Pb isotopes. Meanwhile, such geographic determination in unit (A) would be very uncertain owing to the lack of sediment deposits (possibly due to harbor dredging; Bony et al., 2011; Morhange and Marriner, 2010). Considering the protohistoric period (unit B), we only did take into account Cu mine sources owing to (1) the significant occurrence of Cu artefacts (and the lack of Pb ones) in regional Iron Age settlements (Fiches et al., 1995; Rivet

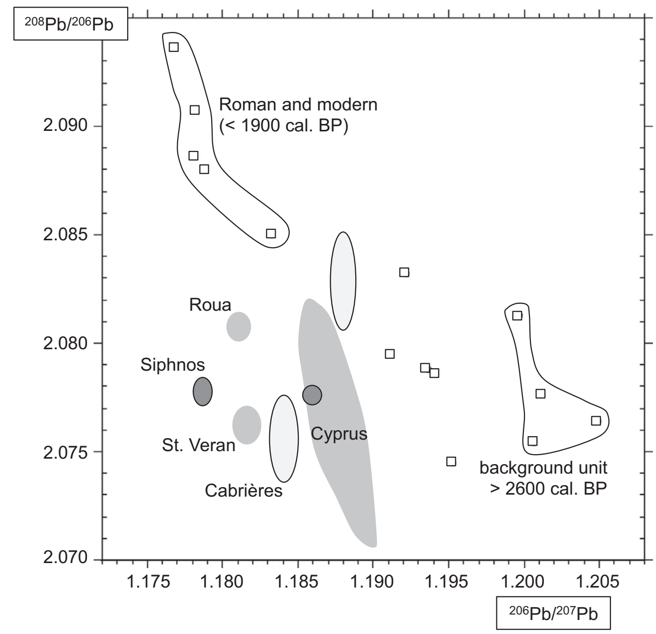

Fig. 4. ^{208}Pb/^{206}Pb versus ^{206}Pb/^{207}Pb isotopic data plot (open squares) with possible matching ore imprints from Cyprus, Alpine, Languedoc (circled light gray) and Cycladic (circled dark grey) geographic sources (see details in text). Note that modern and Roman (< 1900 years cal. BP) and background (> 2600 years cal. BP) deposits are circled opened squares. Other samples belong to the enriched protohistoric unit (B) as described in the text.

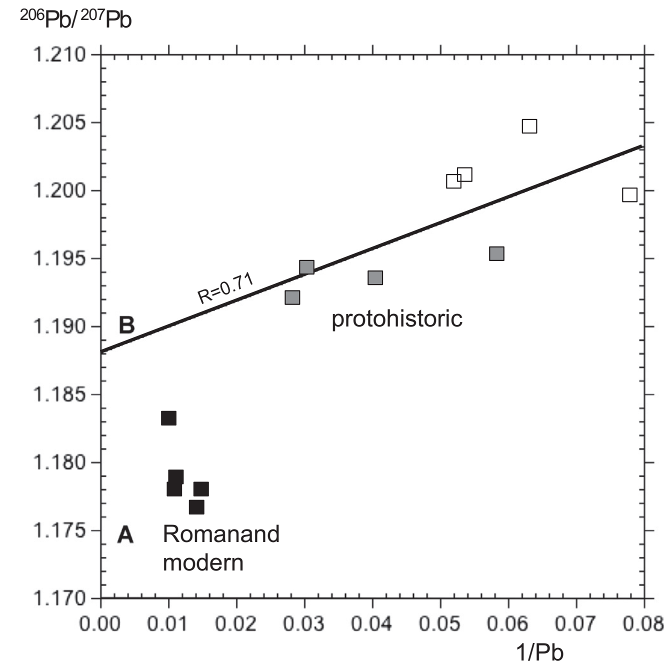

Fig. 5. 1/Pb versus ^{206}Pb/^{207}Pb ratios plot with linear curve fits between the background unit (open squares) and (B) the protohistoric period (circled grey closed squares). The Roman and modern period (A) is shown in dark closed squares.

et al., 2000) and (2) the fact that excess anthropogenic Cu is less than ten times more enriched in the Roman harbor as compared to the protohistoric levels, suggesting the use of Cu forges during the protohistoric period to explain the contamination of the valley to such degree. Pb isotopes allow to investigating the geographical origin of Cu ingots used for bronzes and native Cu-artifacts (Brill and Wampler, 1967; Gale, 1999, 2001; Gale and Stos-Gale, 1982, 1986, 2000; Stos-Gale and Gale, 2009; Knapp, 2000; Scaife, 1997) of which imprints are recorded from Fréjus ancient bay sediments. Isotopic mixing between natural and various ore sources enable to identifying possible geographic origins in ^{206}Pb/^{207}Pb vs. ^{208}Pb/^{206}Pb isotope plots (Fig. 4). Here we show that the most relevant possible Cu mine origin, based on Pb isotopes, for the sediments deposits during the protohistoric period in the marine sand unit (B) is either from the Alps (Roua and Saint Veran), the Languedoc region (Cabrières) (Fig. 1a) and/or the Limni-Solea district of Cyprus and the island of Siphnos (Cyclades) in eastern Mediterranean. The regional Cu mines of Roua and Saint Veran located in the Southern French Alps, about 150 km and 280 km north of Fréjus respectively, are among the mines that comprise easily extractible native Cu in the Alps. Along with Cabrières mines, there are among the very few known metallurgical Cu sites in southern Gaul during the protohistoric period. Their contribution to Cu ores production in the Mediterranean is negligible as compared to that of the Iberian or Cyprus regions. All of these mines in the Alps, the Languedoc, Cyprus and the Cyclades have been quarried at least since the Bronze Age (Ambert, 1999; Bourgarit et al., 2008, 2010; Cattin, 2008; Chalkias et al., 1988; Domergue et al., 2006; Espérou et al., 1994; Gale and Stos-Gale, 1985, 1986, 1999; Gale et al., 1981, 1997; Hamelin et al., 1988; Mari, 2002; Prange and Ambert, 2005; Rostan and Mari, 2005; Spooner and Gale, 1982; Stos-Gale and Gale, 1994; Stos-Gale et al., 1996, 1997; Web et al., 2006) and therefore their ingots could have contributed metal imprints and associated enrichments found in protohistoric sediment deposits at Fréjus. Several uncertainties are associated with the use of Pb isotope ratios to determine the geographic Cu and Pb ore provenance based on Pb isotope ratios that include (1) ore deposit heterogeneity, (2) isotopic fractionation during metallurgical processes and (3) recycling and pooling of re-smelted metal artifacts. Isotopic fractionation on Pb isotope ratios from smelting operation is generally below 0.2% (Barnes et al., 1974; Stos-Gale and Gale, 2009) and therefore shall not impede our conclusions. Unfortunately much fewer data are available for the Alpine and Cycladic Cu mines considered for this study (Cattin, 2008). Possible geographic mine fields do not overcome the protohistoric Fréjus data set (Fig. 4) owing to the contribution of Pb from both Cu ingots and crustal (background) origin. In order to correct this background contribution and verify our isotopic-based provenance determination we combine Pb concentrations and corresponding ^{206}Pb/^{207}Pb ratios as shown in figure 5 where mixing lines are drawn between the geochemical background data points and measurements in unit (B) (protohistoric period):

(B) $y = 1.188 + 0.189x$ with correlation factor $r = 0.71$ where $x = 1/\text{Pb}$ and $y = {}^{206}\text{Pb}/{}^{207}\text{Pb}$

The intersect between linear curve fit (B) and the y-axis (1/Pb) provide the best possible estimate on the mean signature of the anthropogenic sources (and mix thereof) that contributed to the mean ^{206}Pb/^{207}Pb ratio measured in the sediment unit, i.e. ^{206}Pb/^{207}Pb = 1.188 (Fig. 5). We determine a maximum uncertainty of 0.5% on this isotopic ratio by adding the standard deviations of the mean ^{206}Pb/^{207}Pb ratios measured in the sedimentological units. Considering this uncertainty, we calculate an anthropogenic imprint range for unit B = 1.182–1.194 that remains consistent with the considered mine sources with the exception of Siphnos, in the Cyclades (Fig. 4). It should be noted that the distribution of data for unit (A) (Roman and modern) in Fig. 5 suggests multiple source imprints as evoked in the previous paragraph. The Alpine origin for copper ingot is consistent with archaeological findings regarding known mines-related economical exchanges since the Bronze Age in southeastern France (Bailly-Maitre and Gonon, 2008; Cattin, 2008; Leveau, 2016; Pagès, 2008). Furthermore, the few investigated tombs in Fréjus region have provided cultural insights that link protohistoric settlements to other regional and northern Alpine cultures (Bérato et al., 1991; Excoffon et al., 2010a, 2010b). The Languedoc source is geochemically less likely assuming its very low Pb content (generally below 0.01%) that would limit its detection in environmental samples and would not be consistent with the correlation found between Cu and Pb in sediments. Although coherent with isotope imprints and their known polymetallic characteristics, the Cyprus eastern Mediterranean source for protohistoric Cu found at Fréjus remain a chemical assumption that is not supported by any archaeological evidences and therefore should be considered with caution. Further isotopic analyses on regional Cu artefacts may help resolve this issue and possibly bring new insights on metal trades in southern Gaul during the Iron Age.

5. Conclusions

Here we take advantage of the well described and dated paleoenvironmental findings from Fréjus, one of the main Roman harbors in the Western Mediterranean, in order to investigate geochemical imprints associated with the onset of the Roman city and preceding protohistoric occupation. Trace metal concentrations and corresponding isotopic imprints unmistakably show not only the geochemical impact of the development of the harbor and Augustan city, in excellent agreement with other paleoenvironmental findings, but also new protohistoric imprints prevailing the establishment of the Roman colony. Pb isotopes are proven useful to discern human activities during the Iron Age in the Argens delta. Further archaeological and paleoenvironmental investigations are needed to estimate the importance of metallurgical activity during the Iron Age in this area, and most particularly that of the Escaravatiers protohistoric settlement, as well as the dispersion, transport and deposition modes of pollutant metals into local coastal sediments. Pb isotope analyses suggest several geographical ore sources (Alps, Languedoc, and eastern Mediterranean) that may have contributed to pollutant Cu accumulated within protohistoric sediment deposits. The isotopic analysis of copper-made artifacts found in nearby Iron Age settlements should help better ascertain the geographical origin of the ores from which metals were extracted and bring insights into metal forges and trades in the lower Argens valley during the protohistoric period.

Acknowledgments

The project leading to this publication has received partial funding from Excellence Initiative of Aix-Marseille University (A*MIDEX, a French “Investissements d’Avenir” program). The authors thank Fréjus Town Council, The “Service Municipal d'Archéologie” and the “Service Départemental d'Archéologie” for logistic and funding. This research was conducted as part of the Paleomed ANR program. We are grateful to P. Leveau, P. Excoffon and two anonymous reviewers for providing insightful comments that helped improve the manuscript.

References

Ambert, P., 1999. Grandeurs et décadences des exploitations de cuivre de Cabrières (Hérault) entre néolithique et moyen âge. Pallas 50 (1), 391–404.
Angelidis, M.O., Radakovitch, O., Véron, A., Aloupi, M., Heussner, S., Price, B., 2011. Pollutant metal and sapropel imprints in deep Mediterranean sediments. Mar. Pollut. Bull. 62, 1041–1052.
Bailly-Maitre, M.C., Gonon, T., 2008. L'exploitation de la chalcopyrite à l'âge du Bronze dans le massif des Rousses en Oisans (Isère): premiers éléments. In: Richard, H., García, D. (Eds.), Le peuplement de l’arc alpin. CTHS, pp. 207–223 Paris.
Barnes, I.L., Shields, W.R., Murphy, T.J., Brill, R.H., 1974. Isotopic analysis of Laurion lead ores. In: Beck, C.W. (Ed.), Archaeological Chemistry. American Chemical Society, Washington, pp. 1–10.
Bérato, J., Magnin, F., 1989. Le Touar, Les Arcs-sur-Argens (Var). Un habitat de plaine du Bronze final II/IIIa et du premier Âge du fer dans son environnement. Doc. Archéol. Mérid. 12, 7–40.
Bérato, J., Dugas, F., Babillaud, C., 1990. Retru, Trans-en-Provence, habitat de plaine du premier Âge du fer et du Haut Empire. In: A. Soc. Sci. Nat. Archéol. Toulon et Var (Ed.). Travaux du centre archéologique du Var, pp. 452–455.
Bérato, J., Dugas, F., Dutour, O., 1991. Les tombes protohistoriques de Gros-Ped, Les Arcsur-Argens (Var). Doc. Archéol. Méridionale 14 (1), 125–140.
Bertoncello, F., 1999. Le peuplement de la basse vallée de l'Argens et de ses marges (Var, France), de la fin de l'Âge du fer à la fin de l'Antiquité. PhD Thesis. University of Provence, Aix en Provence.
Bertoncello, F., 2005. L'occupation d'un milieu de moyenne montagne en Provence orientale : le massif des Maures (Var) du IIe siècle av. J.-C. au VIIe siècle. In: A. Bouet A. et F. Verdin (Eds.). Territoires et paysages de l’âge du Fer au Moyen Âge, Mélanges offerts à Philippe Leveau, Bordeaux, Ausonius Éditions, pp. 45–61.
Bertoncello, F., Devillers, B., 2006. Une vision renouvelée de la dynamique du paysage dans la basse vallée de l'Argens : synthèse paléo-environnementale préliminaire. ArchéoSciences - Revue d'Archéométrie 30, 223–225.
Bertoncello, F., Bonnet, S., Bouby, L., Delhon, C., Devillers, B., Guillon, S., 2008. Occupation du sol et patrimoine archéologique dans la basse vallée de l'Argens. In: Projet collectif de recherche, Rapport de fin de projet. CNRS-Ministère de la Culture et de la Communication, Valbonne 124 p.
Bertoncello, F., Devillers, B., Bonnet, S., Guillon, S., Bouby, L., Delhon, C., 2014. Mobilité des paysages littoraux et peuplement dans la basse vallée de l'Argens (Var, France) au cours de l'Holocène. Quaternaire 25 (1), 23–44.
Bony, G., Morhange, C., Bruneton, H., Carbonel, P., Gébara, C., 2011. 2000 ans de colmatage du port antique de Fréjus (*Forum Julii*), France: une double métamorphose littorale. C. R. Geosciences 343, 701–715.
Bourgarit, D., Rostan, P., Burger, E., Carozza, L., Artioli, G., 2008. The beginning of copper mass production in the western Alps : the Saint Véran mining area reconsidered. Hist. Metall. 42 (1), 1–11.
Bourgarit, D., Rostan, P., Carozza, L., Mille, B., Artioli, G., 2010. 20 ans de recherches autour du site de St Véran à l'âge du Bronze ancien: état de la question. Trab. Prehist. 67 (2), 269–285.
Brill, R.H., Wampler, J.M., 1967. Isotope studies of ancient lead. Am. J. Archaeol. 71, 63–77.
Brun, J.P., 1999. Carte archéologique de la Gaule 83/1 et 83/2: le Var. Académie des Inscriptions et Belles-Lettres, Paris 984 p.
Cattin, F., 2008. Modalité d’approvisionnement et modalités de consommation du cuivre dans les Alpes au 3eme millénaire avant notre ère: apport des analyses métalliques à la connaissance des peuplements du Néolithique final, du Campaniforme et du Bronze ancien. Ph.D. thesis. University of Geneva 458 p.
Chalkias, S., Vavelidis, M., Schmitt-Strecker, S., Begemann, F., 1988. Geologische Interpretation der Blei-isotopenverha Èltnisse von Erzen der Insel Thasos, derAÈ gaÈis und Nordgriechenlands. In: Wagner, G.A., Weisgerber, G. (Eds.), Antike Edel und Buntmetallgewinnung auf Thasos Der Anschnitt, Zeitschrift fur Kunst und Kultur im Bergau, Supplement 6, Deutches Bergaumuseum, Bochum, pp. 59–74.
Chevallier, R., 1998. Les voies romaines. Picard Editions, Paris 343 p.
Christol, M., 2015. Pacensis: les noms de la cité de Fréjus et l'histoire coloniale sous Auguste. Mélanges de l'École Française de Rome 172 (2), 535–553.
De Vleeschouwer, F., Gerard, F., Goormaghtigh, C., Mattielli, N., Le Roux, G., Fagel, N., 2007. Last two millennia atmospheric lead and heavy metals inputs in a Belgian peat bog: regional to global human impacts. Sci. Tot. Env. 377, 297–310.
De Vleeschouwer, F., Fagel, N., Cheburkin, A., Pazdur, A., Sikorski, J., Mattielli, N., Renson, V., Fialkiewicz, B., Piotrowska, N., 2009. Anthropogenic impacts in North Poland over the last 1300 years. Sci. Tot. Env. 407, 5674–5684.
Delile, H., Mazzini, I., Blichert-Toft, J., Goiran, J.P., Arnaud-Godet, F., Salomon, F., Albarede, F., 2014. Geochemical investigation of a sediment core from the Trajan basin at Portus, the harbor of ancient Rome. Quat. Sci. Revue 87, 34–45.
Devillers, B., Bonnet, S., 2006. 6000 ans d'histoire environnementale de la lagune de Villepey (Fréjus, Var): premiers résultats de l'étude multi-proxies de la carotte Vil2. Archéosciences - Revue d'Archéométrie 30, 197–203.
Devillers, B., Excoffon, P., Morhange, C., Bonnet, S., Bertoncello, F., 2007. Relative sea-level changes and coastal evolution at Forum Julii (Frejus, Provence. C. R. Geosci. 339 (5), 329–336.
Doe, B.R., 1970. Lead Isotopes. Springer Verlag, Berlin, Heidelberg, New York.
Domergue, C., Serneels, V., Cauuet, B., Paillet, J.M., Orzechowski, M., 2006. Mines et métallurgies en Gaule à la fin de l'âge du Fer et à l’époque romaine. Celtes et Gaulois, l'Archéologie face à l'Histoire, 5: la romanisation et la question de l’héritage celtique. In: Actes de la table ronde de Lausanne, 17–18 juin. 2005, Bibracte, Centre archéologique européen, Glux-en-Glenne, pp. 131–162.
Doucelance, R., Manhès, G., 2001. Reevaluation of precise lead isotope measurements by thermal ionization mass spectrometry: comparison with determinations by plasma source mass spectrometry. Chem. Geol. 176, 361–377.
Dubar, M., 2004. L'édification deltaïque du bas Argens (Var, France) durant la protohistoire et l'Antiquité. Application d'un modèle numérique 2D à l'archéologie. Méditerranée 102, 47–53.
Duval, P.M., 1971. La Gaule jusqu'au milieu du V^{e} siècle (Les sources de l'histoire de France des origines à la fin du XV^{e} siècle, Paris).
Elmaleh, A., Galy, A., Allard, T., Dairon, R., Day, J.A., Michel, F., Marriner, N., Morhange, C., Couffignal, F., 2012. Anthropogenic accumulation of metals and metalloids in carbonate-rich sediments : insights from the ancient harbor setting of Tyre (Lebanon). Geochim. Cosmochim. Acta 82, 23–38.
Espérou, J.L., Ambert, P., Bourhis, J.R., Roques, P., Gilot, E., Chabal, L., 1994. La fosse chalcolithique Pioch-Farrus 448 (Cabrières, Hérault): datation ^{14}C et documents métallurgiques. Bull. du musée d'anthropol. préhist. Monaco 37, 53–62.
Excoffon, P., Devillers, B., Bonnet, S., Boudy, L., 2006. Nouvelles données sur la position du littoral de Fréjus. Le diagnostic archéologique du “théâtre d'agglomération” (Fréjus, Var). ArchéoSciences - Revue d'Archéométrie 30, 205–211.
Excoffon, P., Bailet, P., Henry, A., Rodet-Belarbi, I., Pâques, J., 2010a. Une tombe à incinération du premier âge du Fer au quartier du Capitou à Fréjus (Var). Bulletin Archéo. de Provence 33, 19–26.
Excoffon, P., Bailet, P., La Rocca, C., 2010b. Le quartier du théâtre antique: des traces d'occupation protohistorique et quelques données topographiques nouvelles sur la ville romaine (Fréjus, Var). Revue du Centre Archéo. du Var 205–219.
Excoffon, P., Bonnet, S., Tomatis, C., Ferrer, L., Ardisson, S., Bernigaud, N., Portalier, N., Morhange, C., 2016. Restitution de la morphologie littorale et aménagements portuaires à *Forum Iulii*. In: Sanchez, C., Jézégou, M.P. (Eds.), Actes du colloque: les ports

dans l'espace méditerranéen antique. 44e Supplément à la Revue Archéologique de Narbonnaise, pp. 1–14.
Ferrand, J.L., Hamelin, B., Monaco, A., 1999. Isotopic tracing of anthropogenic Pb inventories and sedimentary fluxes in the Gulf of Lions (NW Mediterranean sea). Cont. Shelf Res. 19, 23–47.
Fiches, J.L., Brentchaloff, D., Chouquer, G., Dubar, M., Gazenbeek, M., Latour, J., Rogers, G.B., 1995. Habitats de l'Âge du Fer et structures agraires d'époque romaine aux Escaravatiers (Puget-sur-Argens, Var). Gallia 52, 205–261.
Gale, N.H., 1999. Lead isotope characterization of the ore deposits of Cyprus and Sardinia and its application to the discovery of the sources of copper for Late Bronze Age oxhide ingots. In: Young, S.M.M., Pollard, A.M., Budd, P., Ixer, R.A. (Eds.), Metals in Antiquity, British Archaeological Reports, International Series. Vol. 792. Archaeopress, Oxford, pp. 110–121.
Gale, N.H., 2001. Archaeology, science-based archaeology and the Mediterranean Bronze Age metals trade : a contribution to the debate. Eur. J. Archaeol. 4 (1), 113–130.
Gale, N.H., Stos-Gale, Z.A., 1982. Bronze and copper sources in the Mediterranean : a new approach. Science 216, 11–19.
Gale, N.H., Stos-Gale, Z.A., 1985. Cyprus and the Bronze Age metals trade. In: Proceedings of the Second International Congress of Cypriot Studies, Nicosia 1982, pp. 51–66.
Gale, N.H., Stos-Gale, Z.A., 1986. Oxhide ingots in Crete and Cyprus and the Bronze Age metals trade. Annu. Br. Sch. Athens 81, 81–100.
Gale, N.H., Stos-Gale, Z.A., 1999. Copper oxhide ingots and the Aegean metals trade. New perspectives. In: Betancourt, P., Karageorghis, V., Laffineur, R., Niemeyer, W.D. (Eds.), Aegeum 20. Meletemata Studies in Aegean Archaeology, pp. 267–278.
Gale, N.H., Stos-Gale, Z.A., 2000. Lead isotope analyses applied to provenance studies. In: Ciliberto, E., Spoto, G. (Eds.), Modern Analytical Methods in Art and Archaeology, Chemical Analyses Series. 155. John Wiley and Sons, Inc., New-York, pp. 503–584 17.
Gale, N.H., Spooner, E.T.C., Potts, P.J., 1981. The lead and strontium isotope geochemistry of metalliferous sediments associated with Upper Cretaceous ophiolitic rocks in Cyprus, Syria, and the Sultanate of Oman. Can. J. Earth Sci. 18, 1290–1302.
Gale, N.H., Gale, Z.A., Maliotis, G., Annetts, N., 1997. Lead isotope data from the isotrace Laboratory, Oxford: archaeometry data base 4, ores from Cyprus. Archaeometry 39 (1), 237–246.
Gascou, J., 1982. Quand la colonie de Fréjus fut-elle fondée? Latomus 41 (1), 132–145.
Gascou, J., Janon, M., 1985. Inscriptions latines de Narbonnaise (*ILN Fréjus*). Gallia suppl. 44 (Paris).
Gébara, C., 2012. Carte archéologique de la Gaule 83/3 : Fréjus. Académie des Inscriptions et Belles-Lettres, Paris 513 p.
Gébara, C., Morhange, C., 2010. Fréjus (*Forum Iulii*): le port antique/The ancient harbour. J. Roman Archaeol. 152 Suppl. Series 77.
Hamelin, B., Dupre, B., Brevart, O., Allegre, C.J., 1988. Metallogenesis at Paleo-spreading centers : lead isotopes in sulfides, rocks and sediments from the Troodos ophiolite (Cyprus). Chem. Geol. 68, 229–238.
Huisman, D.J., Vermeulen, F.J.H., Baker, J., Veldkamp, A., Kroonenberg, S.B., Klaver, G.Th, 1997. A geological interpretation of heavy metal concentrations in soils and sediments in the southern Netherlands. J. Geochem. Explor. 59, 163–174.
Knapp, A.B., 2000. Archaeology, science-based archaeology and the Mediterranean Bronze Age metals trade 2000. Eur. J. Archaeol. 3 (1), 31–56.
Le Roux, G., Veron, A., Morhange, C., 2003. Geochemical evidences of early anthropogenic activity in harbour sediments from Sidon. Archeol. Hist. Leb 18, 115–119.
Le Roux, G., Morhange, C., Veron, A., 2005. Lead pollution in the ancient harbor of Marseilles. Méditerranée 1 (2), 31–35.
Leveau, P., 2016. La circulation des biens et des Hommes dans les Alpes entre la préhistoire et le moyen âge : remarques conclusives. In: Proceedings of XIVth Colloque sur les Alpes dans l'Antiquité Evolène/Valais, Switzerland, 2–4 Oct. 2015, Bulletin d'Études Préhistoriques et Archéologiques Alpines, pp. 9–14.
Manhès, G., Minster, J.F., Allegre, C.J., 1978. Comparative uranium-thorium-lead and rubidium-strontium study of the Saint Severin amphoterite: consequences for early solar system chronology. Earth Planet. Sci. Lett. 39, 14–24.
Mari, G., 2002. Les gîtes cuprifères du Dôme de Barrot (Alpes-Maritimes, France). In: Bull. d'études préhist. et archeol. alpines (Aoste) 13, Colloque sur les Alpes dans l'Antiquité (Tende, 15–17 Sept. 2000), pp. 13–26.
Miralles, J., Veron, A.J., Radakovitch, O., Deschamps, P., Tremblay, T., Hamelin, B., 2006. Atmospheric lead fallout over the last century recorded in Gulf of Lions sediments (Mediterranean Sea). Mar. Pollut. Bull. 52, 1364–1371.
Morhange, C., Marriner, N., 2010. Roman dredging in ancient Mediterranean harbours. Bulletino di Archeologia Online vol. spec. IAAC. http://151.12.58.75/archeologia/bao_document/articoli/4_Morhange_Marriner_paper.pdf.
Pagès, G., 2008. La métallurgie du fer en France méditerranéenne de l'Antiquité au début du Moyen Âge : jalons d'une approche interdisciplinaire. PhD Thesis. University Montpellier III - Paul Valéry 265 p.
Pasqualini, M., 2011. Fréjus: Forum Iulii Octavanorum colonia, quae Pacensis appellatur et classica. In: Pasqualini (Ed.), Fréjus romaine, la ville et son territoire. Agglomérations de Narbonnaise, des Alpes-Maritimes et de Cisalpine à travers la recherche archéologique. Actes du 8e colloque historique de Fréjus, 8–10 Octobre 2010, pp. 15–52 Editions APDCA, Antibes.
Pellissier, M., 2006. Le Muy, Les Vaugreniers. Bilan Scientifique. In: Direction Régionale des Affaires Culturelles Provence-Alpes-Côted'Azur, Service Régional de l'Archéologie, pp. 171–172.
Philip, G., Clogg, P.W., Dungworth, D., 2003. Copper metallurgy in the Jordan Valley from the third to the first millennia BC: chemical, metallographic and lead isotope analyses of artefacts from Pella. Levant 33, 71–100.
Prange, M., Ambert, P., 2005. Caractérisation géochimique et isotopique des minerais et des métaux base cuivre de Cabrières (Hérault). In: Ambert, P., Vaquer, J. (Eds.), La première métallurgie en France et dans les pays limitrophes, Actes du colloque international Carcassonne, 28–30 Sept. 2002, Soc. Prehist. Fr., Paris, mémoire. 37. pp. 71–81.
Provansal, M., Bertoncello, F., Gili, G., 2006. Dynamique morpho-sédimentaire et peuplement sur le Rocher de Roquebrune-sur-Argens (Var). ArchéoSciences - Revue d'Archéométrie 30, 173–180.
Py, V., Veron, A., Edouard, J.L., de Beaulieu, J.L., Ancel, B., Segard, M., Durand, A., Leveau, P., 2014. Interdisciplinary characterisation and environmental imprints of mining and forestry in the upper Durance valley (France) during the Holocene. Quat. Int. 353, 74–97.
Rivet, L., Brentchaloff, D., Roucole, S., Saulnier, S., 2000. Atlas topographique des villes de Gaule méridionale 2 : Revue archéologique de Narbonnaise. 32 509 p.
Rostan, P., Mari, G., 2005. L'exploitation protohistorique de cuivre natif de Roua (Daluis et Guillaumes, Alpes-Maritimes). In: Ambert, P., Vacquer, J. (Eds.), Actes de la conférence sur la première métallurgie en France et dans les pays limitrophes (Carcassonne 2002). Société Préhistorique Française, pp. 139–149 Paris.
Roussiez, V., Ludwig, W., Probst, J.L., Monaco, A., 2005. Background levels of heavy metals in surficial sediments of the Gulf of Lions (NW Mediterranean): an approach based on 133Cs normalization and lead isotope measurements. Env. Poll. 138, 167–177.
Sayre, E.V., Joel, E.C., Blackman, M.J., Yener, K.A., Ozbal, O., 2001. Stable lead isotope studies of Black Sea Anatolian ore sources and related Bronze Age and Phrygian artefacts from nearby archaeological sites, Appendix: new Central Taurus ores data. Archaeometry 43 (1), 77–115.
Scaife, B., 1997. Database of lead isotope ratios for ores collected from around the Mediterranean. Papers Published between 1987 and 1997. http://www.brettscaife.net/lead/data/index.html.
Shotyk, W., Krachler, M., Martinez-Cortizas, A., Cheburkin, A.K., Emons, H., 2002. A peat bog record of natural, pre-anthropogenic enrichments of trace elements in atmospheric aerosols since 12370 14C yr, and their variation with Holocene climate change. Earth Planet. Sci. Lett. 199, 21–37.
Spooner, E.T.C., Gale, N.H., 1982. Pb isotopic composition of ophiolitic volcanogenic sulphide deposits, Troodos Complex, Cyprus. Nature 296, 239–242.
Stanley, J.D., Carlson, R.W., Van Beek, G., Jorstad, T.F., Landau, E.A., 2007. Alexandria, Egypt, before Alexander the Great: a multidisciplinary approach yields rich discoveries. GSA Today 17 (8), 4–10.
Stos-Gale, Z.A., Gale, N.H., 1994. The origin of metals excavated on Cyprus. In: Knapp, B., Cherry, J. (Eds.), Provenance Studies and Bronze Age Cyprus: Production Exchange and Politico-economic Change. vol. 3 Prehistory Press, Madison pp. 92–122 and 210–216.
Stos-Gale, Z.A., Gale, N.H., 2009. Metal provenancing using isotopes and the Oxford archaeological lead isotope database (OXALID). Archaeol. Anthropol. Sci. 1 (3), 195–213.
Stos-Gale, Z.A., Gale, N.H., Annetts, N., 1996. Lead isotope data from the isotrace laboratory, Oxford: archaeometry data base 3, ores from the Aegean, part 1. Archaeometry 38, 381–390.
Stos-Gale, Z.A., Maliotis, G., Gale, N.H., Annetts, N., 1997. Lead isotope characteristics of the Cyprus copper ore deposits applied to provenance studies of copper oxhide ingots. Archaeometry 39 (1), 83–124.
Véron, A., Goiran, J.P., Morhange, C., Marriner, N., Empereur, J.Y., 2006. Pollutant lead reveals the pre-Hellenistic occupation and ancient growth of Alexandria, Egypt. Geophys. Res. Lett. 33, 1–4.
Véron, A., Le Roux, G., Poirier, A., Braque, D., 2012. Origin of copper used in bronze artifacts from Middle Bronze Age burials in Sidon: a synthesis from lead isotope imprint and chemical analyses. Archeol. Hist. Leb 34-35, 68–78.
Veron, A., Flaux, C., Marriner, N., Poirier, A., Rigaud, S., Morhange, C., Empereur, J.Y., 2013. A 6000-year geochemical record of human activities from Alexandria (Egypt). Quat. Sci. Rev. 81, 138–147.
Web, J.M., Frankel, D., Stos-Gale, Z.A., Gale, N.H., 2006. Early Bronze Age metal trade in the eastern Mediterranean : new compositional and lead isotope evidence from Cyprus. Oxf. J. Archaeol. 25 (3), 261–288.