

# The economic resilience of Carthage during the Punic Wars: insights from sediments of the Medjerda delta around Utica (Tunisia)

Hugo Delile<sup>1</sup>, Elisa Pleuger<sup>2</sup>, Janne Blichert-Toft<sup>3</sup>, Jean-Philippe Goiran<sup>4</sup>, Nathalie Fagel<sup>5</sup>, Ahmed Gadhoum<sup>6</sup>, Hakim Abichou<sup>7</sup>, Imed Ben Jerbania<sup>8</sup>, Elizabeth Fentress<sup>9</sup>, Andrew Wilson<sup>10</sup>

<sup>1</sup>Maison de l'Orient et de la Méditerranée, CNRS UMR 5133, 69365 Lyon Cedex 7, France France, <sup>2</sup>Département de Géologie, UR Argiles, Géochimie et Environnements Sédimentaires (AGEs), Université de Liège, Liège, Belgium, <sup>3</sup>Ecole Normale Supérieure de Lyon, <sup>4</sup>CNRS - UMR 5133 Archéorient, <sup>5</sup>Département de Géologie, UR Argiles, Géochimie et Environnements Sédimentaires (AGEs), Université de Liège, Liège, Belgium, <sup>6</sup>Département d'Archéologie Sous-Marine, Institut National du Patrimoine, Tunis, Tunisia, <sup>7</sup>Laboratoire de Cartographie Géomorphologique des Milieux, des Environnements et des Dynamiques, Faculté des Sciences Humaines et Sociales de Tunis, Tunis, Tunisia, <sup>8</sup>Institut National du Patrimoine, Tunis, Tunisia, <sup>9</sup>Independent scholar, <sup>10</sup>University of Oxford

Submitted to Proceedings of the National Academy of Sciences of the United States of America

**While the Punic Wars (264–146 BC) have been the subject of numerous studies, generally focused on their most sensational aspects (major battles, techniques of warfare, geopolitical strategies, etc.), curiously, the exceptional economic resilience of the Carthaginians in the face of successive defeats, loss of mining territory, and the imposition of war reparations has attracted hardly any attention. Here we address this issue using a newly developed powerful tracer in geoarchaeology, that of Pb isotopes applied to palaeo-pollution. We measured the Pb isotopic compositions of a well-dated suite of eight deep cores taken in the Medjerda delta around the city of Utica. The data provide the first robust evidence of ancient lead-silver mining in Tunisia and lay out a chronology for its exploitation which appears to follow the main periods of geopolitical instability at the time: the Greco-Punic Wars (480–307 BC) and the Punic Wars (264–146 BC). During the last conflict, the data further suggest that Carthage was still able to pay indemnities and fund armies despite the loss of its traditional silver sources in the Mediterranean. This work shows that the mining of Tunisian metalliferous ores between the second half of the 4<sup>th</sup> and the beginning of the 3<sup>rd</sup> c. BC contributed to the emergence of Punic coinage and the development of the Carthaginian economy.**

palaeo-pollution | mining resources | Medjerda river | Punic Wars | Utica

## Introduction

The three Punic wars (264–146 BC) saw two of the greatest empires of antiquity struggle for over a century for control of the western Mediterranean: Punic Carthage, a maritime power whose territory at the dawn of the First Punic War (264–241 BC) formed a narrow fringe along the coasts of North Africa and those of Andalusia, including the islands of the western Mediterranean; and Rome, an emerging terrestrial power on the Italian peninsula. Our knowledge of this conflict is dependent on the nature of the available sources, principally historical records, and, secondarily, on epigraphy, numismatics and archaeology (1,2). Each of these disciplines deals with complementary aspects covering various fields such as the techniques of warfare employed, the geopolitical strategies implemented, or the resources committed. A particular difficulty when dealing with the ancient historical accounts (2) is source bias: no Punic texts have been preserved, and the Latin authors showed a notable bias against their Punic enemies (1).

A key problem concerns the financial and monetary means used to support the war effort, which depended on access to mining resources, especially lead-silver ores, critical for ancient economies (3–5). Our knowledge of ancient mining centres (i) is focused mainly on Roman mining districts (a question of textual sources?) (6, 7), (ii) tends to emphasize major centres to the detriment of smaller ones (8) and (iii) concerns Central

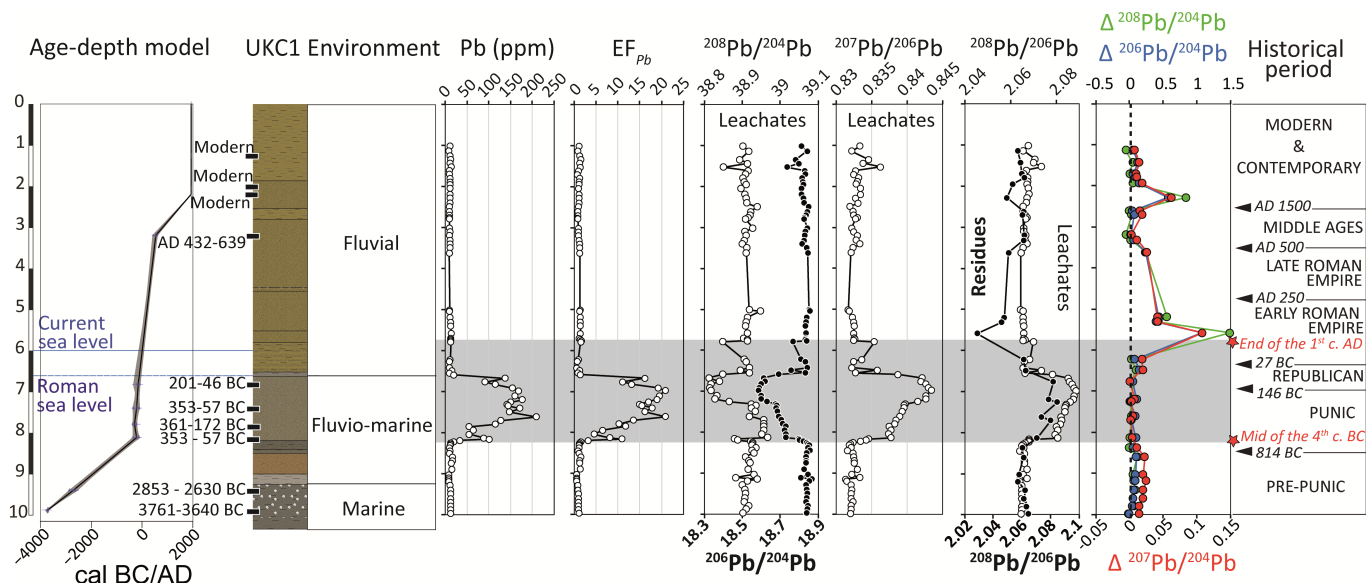
and Western Europe more than North Africa (5–7, 9–12), often without robust chronological timelines (7). This paradox is all the more striking when one observes the spatial distribution of lead-silver mines within the Roman Empire, which is largely devoid of them along its southern and eastern fringes (5–7). Information on the large mining regions exploited by the Carthaginians is limited to southern Spain (5, 6), Sardinia and Sicily (13) territories which were annexed by the Romans during, respectively, the 1<sup>st</sup> (264–241 BC) and the 2<sup>nd</sup> Punic War (218–201 BC). What, then, was the geographical origin of the metalresources of the Carthaginians that supported the war effort during the later Punic Wars? Shedding new light on this underestimated aspect is required to put the exceptional resilience of the Carthaginians against the Romans into perspective. Despite the large amount of evidence accumulated over the last two centuries for the possible exploitation of metal resources in Tunisia since antiquity (14–17), mining archaeology has not yet demonstrated that lead/silver exploitation actually took place, still less established a chronology. Only one historical account may refer to the mines of North Africa, a letter of Saint Cyprian dated AD 257 (*Ep.* 76) (18), which mentions Christian convicts sent to mines, but without any useful details on either the nature or the location of the mines in question (14, 15).

## Significance

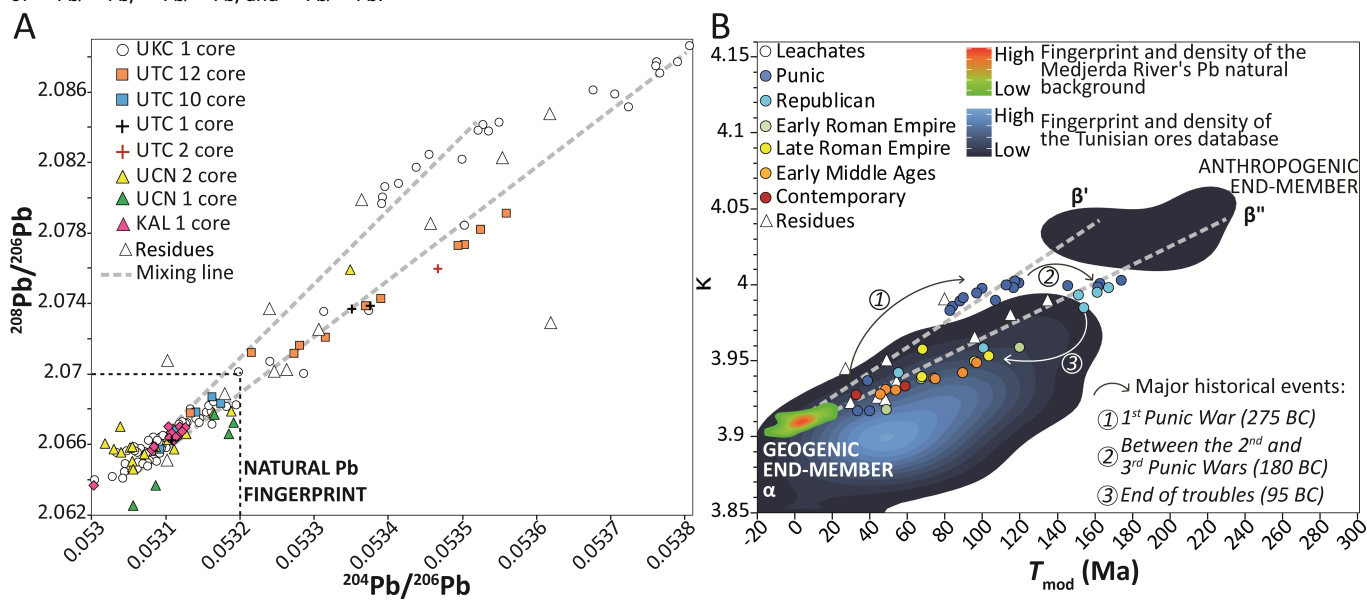
**How do we explain the exceptional economic resilience for more than a century and a half of the Carthaginian civilization against the Romans during the Punic Wars? Based on eight deep cores taken in the Medjerda delta around the city of Utica in Tunisia, we show that the sustainable retreat of Carthage into its hinterland during this period of warfare provided the metal resources whose exploitation by the Carthaginians was sufficient to resist the Romans for said long period. The earliest phase of mining activity recorded in the Utica sediments occurred during the Greco-Punic Wars (480–307 BC) and is coeval with the first minting of Punic coins at Carthage, from which point on the Carthaginian economy became increasingly monetized.**

## Reserved for Publication Footnotes

137  
138  
139  
140  
141  
142  
143  
144  
145  
146  
147  
148  
149  
150  
151  
152  
153  
154  
155  
156  
157  
158  
159  
160  
161  
162  
163  
164  
165  
166  
167  
168  
169  
170  
171  
172  
173  
174  
175  
176  
177  
178  
179  
180  
181  
182  
183  
184  
185  
186  
187  
188  
189  
190  
191  
192  
193  
194  
195  
196  
197  
198  
199  
200  
201  
202  
203  
204



**Fig. 1.** Stratigraphic log of reference core UKC1 showing the palaeo-environmental successions, current (22) and Roman (48) sea levels, and the  $^{14}\text{C}$  age-depth model of core UKC1 constructed using the Clam software (47) on 10 radiocarbon dates (shown with black labels on the stratigraphic log). From this age-depth model are derived both the historical time slice boundaries (indicated with black arrowheads) and the time interval of anthropogenic lead pollution highlighted by the gray band and red stars. This figure shows the Pb concentrations (in ppm), the Pb Enrichment Factor ( $EF_{Pb}$ ), downcore variations of  $^{208}\text{Pb}/^{204}\text{Pb}$ ,  $^{206}\text{Pb}/^{204}\text{Pb}$ ,  $^{207}\text{Pb}/^{206}\text{Pb}$ , and  $^{208}\text{Pb}/^{206}\text{Pb}$  for leachates,  $^{208}\text{Pb}/^{206}\text{Pb}$  for residues, and  $\Delta\text{Pb}$  (the isotopic contrast between residue and leachate) of  $^{208}\text{Pb}/^{204}\text{Pb}$ ,  $^{206}\text{Pb}/^{204}\text{Pb}$ , and  $^{207}\text{Pb}/^{204}\text{Pb}$ .



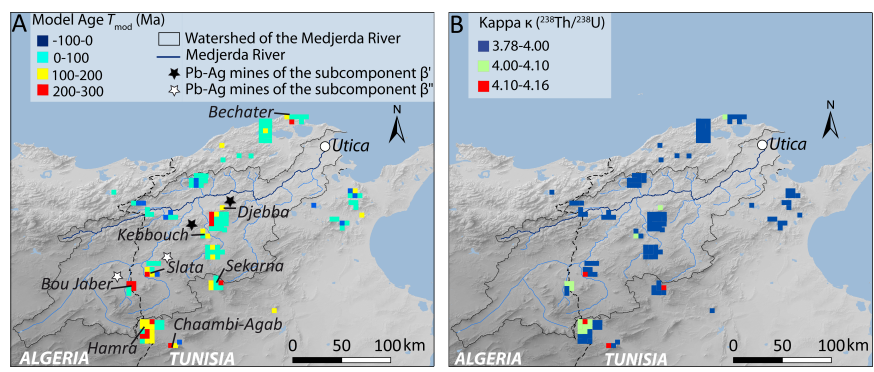
**Fig. 2.** (A) Plot of  $^{204}\text{Pb}/^{206}\text{Pb}$  and  $^{208}\text{Pb}/^{206}\text{Pb}$  for leachates from all eight cores for residues (white triangles) from cores UKC1 and UTC12. (B) Similar plot using the geochemically informed parameters  $T_{\text{mod}}$  and  $k$  for leachates (colored circles) from all eight cores for residues (white triangles) from UKC1 and UTC12. Colors of leachates refer to the major historical periods. The two mixing lines (gray dashes) connect  $\alpha$  and  $\beta'$ , and  $\alpha$  and  $\beta''$ . The  $\alpha$  end-member corresponds to the local geogenic Pb fingerprint (unpolluted water), whereas the  $\beta$  end-member corresponds to the anthropogenic component which in turn exhibits two distinct Tunisian Pb-Ag mining clusters  $\beta'$  and  $\beta''$ . References for the Pb isotope database ( $n = 163$ ) of the Tunisian ores are given in the SI Text.

Here we suggest that the sources normally used to study the Punic Wars (history, epigraphy, numismatics, archaeology, archaeometry) might benefit from being supplemented by geochemistry through the study of Pb palaeo-pollution trapped in environmental records. It is well known that environmental archives, such as the polar icecaps, trapped atmospheric aerosols over several millennia (5), and that high-altitude glaciers (19) and peat bogs (12), as well as lakes (20) at high and medium latitudes, record the signal of past human activity in mining and smelting lead-silver ores. However, there is ongoing debate over the long-distance transport of Pb from its sources (the Pb-Ag mining

districts) into these natural archives, with the spatial extent of the recorded signals ranging from a macro-regional, even hemispheric, scale (5) to a local and/or regional scale, implying strong spatial and temporal variability (12). At much smaller local scales, such as the area of a city, the Pb palaeo-pollution approach is less ambiguous about the source since the distances traveled by anthropogenic lead pollution are short and the routes well established; they follow the urban hydrographic network. Studies of Pb palaeo-pollution trapped in fluvio-marine sediments of ancient harbor basins have thus been able to document unexpected and intriguing facets of urban history in a number of ancient cities

205  
206  
207  
208  
209  
210  
211  
212  
213  
214  
215  
216  
217  
218  
219  
220  
221  
222  
223  
224  
225  
226  
227  
228  
229  
230  
231  
232  
233  
234  
235  
236  
237  
238  
239  
240  
241  
242  
243  
244  
245  
246  
247  
248  
249  
250  
251  
252  
253  
254  
255  
256  
257  
258  
259  
260  
261  
262  
263  
264  
265  
266  
267  
268  
269  
270  
271  
272

273  
274  
275  
276  
277  
278  
279  
280  
281  
282  
283  
284  
285  
286  
287  
288  
289  
290  
291  
292  
293  
294  
295  
296  
297  
298  
299  
300  
301  
302  
303  
304  
305  
306  
307  
308  
309  
310  
311  
312  
313  
314  
315  
316  
317  
318  
319  
320  
321  
322  
323  
324  
325  
326  
327  
328  
329  
330  
331  
332  
333  
334  
335  
336  
337  
338  
339  
340



**Fig. 3.** Map of the Pb isotope database ( $n = 135$ ; see SI Text for references) of Tunisian metal ores (mainly galena) represented as a function of the  $T_{mod}$  (A) and  $\kappa$  (B) parameters. The Pb-Ag mining districts shown with black and white stars (panel A) caused the Pb pollution recorded in the sediment cores because they (i) form the sub-components  $\beta'$  and  $\beta''$  (Fig. 3), respectively, and (ii) are located inside the Medjerda catchment (unlike the mining districts of Hamra, Chaambi-Agab and Sekarna whose Pb isotopic signatures also are similar to those of component  $\beta$ ). The cores of the present study are located around Utica, (white circle).

341  
342  
343  
344  
345  
346  
347  
348  
349  
350  
351  
352  
353  
354  
355  
356  
357  
358  
359  
360  
361  
362  
363  
364  
365  
366  
367  
368  
369  
370  
371  
372  
373  
374  
375  
376  
377  
378  
379  
380  
381  
382  
383  
384  
385  
386  
387  
388  
389  
390  
391  
392  
393  
394  
395  
396  
397  
398  
399  
400  
401  
402  
403  
404  
405  
406  
407  
408

around the Mediterranean (10, 11, 21). These metal palaeo-pollutions trapped in harbor sediments could not have come from aerosols as these contain very low levels of lead, even during episodes of intense atmospheric pollution. For example, during the Roman period when lead contamination of the atmosphere was widespread, the proportion of pollutant lead did not exceed  $\sim 10$  ppm in European lake deposits (20) and a few ppt in Greenland ice (5), while at the same time port sediments were several hundred ppm above the natural Pb abundance level (11). The difference—an order of magnitude or more—is far too large to consider aerosols as a credible source of lead contamination of fluvio-marine sediments. This general pattern of long-range pollutant transport resulting in low lead accumulation must be balanced by the local metal aerosol emissions from mining and metallurgical pollution sources towards areas in their immediate vicinity. Environments near ancient metal-related activity areas could experience Pb enrichments of several tens of ppm during Roman and medieval times (12).

To explore the means used by the Carthaginians to support the war effort during the Punic Wars, we measured the Pb isotopic compositions and concentrations (Dataset S1) of eight sediment cores (UKC1; UCN1, 2; UTC1, 2, 10, 12; and KAL1) taken around the city of *Utica* in the northern part of the Medjerda delta in Tunisia (Fig. S1). UKC1 was selected as the reference core because its position at the exit of the bottleneck between the *Utica* promontory and the hill of *Castra Corneliana* (Fig. S1) means that it carries a record of both the Medjerda watershed and the city of *Utica*. Moreover, the number and quality of  $^{14}\text{C}$  dates measured on the UKC1 core is higher than for the other boreholes.

The Medjerda delta is of particular interest because *Utica* is the oldest Phoenician foundation in the western Mediterranean, with a traditional foundation date of 1101 BC, predating Carthage, although the earliest known archaeological remains are from the 8<sup>th</sup> c. BC. The cores were previously described and dated (Dataset S2) as part of a major geoarchaeological research program conducted over a period of eight years by an international team from Tunisia, Belgium, Britain and France (22, 23).

**Results and Discussion**

**The natural Pb source**

Of the Pb concentrations measured for 146 samples from the eight cores of this study (Fig. S1), 77, or just over half, are considered natural since their Pb enrichment factors ( $EF_{Pb}$ ) are less than 2 (Dataset S1). These samples have relatively low Pb concentrations (an example is shown for the UKC1 core in Fig. 1), with a mean value of 12 ppm, consistent within error bars with the average Pb concentration of 17 ppm for the upper crust (24).

Compared to Pb abundances, Pb isotopes are more suitable for identifying uncontaminated sediments because they are independent of the prevailing environmental conditions (25). First,

the residual fraction left over after leaching a sample in the laboratory corresponds to the Pb contained initially in the mineral crystal lattice during rock formation and hence is referred to as natural, crustal, or detrital since it characterizes the local geogenic Pb background (26). Secondly, the leachate fraction (or extractable phase) represents the labile or anthropogenic component of the sample Pb, which is adsorbed onto sediments once released from an independent anthropogenic source (11, 26). This anthropogenic component (e.g. lead pipes, gasoline, coal, aerosols, etc.) involves “imported” or “exotic” lead whose Pb isotopic composition is often very different from the local geogenic Pb. This is why leachate and residue Pb isotopic compositions are theoretically, and usually in practice as well, distinct in the case of a pure Pb source (e.g. lead pipes). This theoretical framework is in reality more complex because the leached Pb fraction also incorporates part of the natural signal in variable amounts. Consequently, the leached Pb fraction is a mixture between the natural and anthropogenic components, whose respective proportions vary from one sample to another, and the leachates, when plotted in a binary diagram of  $^{204}\text{Pb}/^{206}\text{Pb}$  vs.  $^{208}\text{Pb}/^{206}\text{Pb}$ , form mixing lines between a natural and an anthropogenic source (Fig. 2A). In the case of the Medjerda delta sediments, the isotopic fingerprint of the natural Pb ranges between 0.0530 and 0.0532 and 2.062 and 2.070 for  $^{204}\text{Pb}/^{206}\text{Pb}$  and  $^{208}\text{Pb}/^{206}\text{Pb}$ , respectively (Fig. 2A). This natural Pb isotopic composition is also easily readable using the geochemically informed parameters  $T_{mod}$  (Pb model ages in million years, Ma) and  $\kappa$  (kappa, the  $^{232}\text{Th}/^{238}\text{U}$  ratio), which are derived from the time-integrated Pb isotopic compositions using the equations given by Albarède et al. (27). The advantages of this representation over that based on raw Pb isotopic ratios have been demonstrated in a number of geological (28), archaeological (27) and geoarchaeological contexts (11). The natural Pb isotopic signatures of the Medjerda delta sediments cluster between -20 and 20 Ma and 3.90 and 3.92 for  $T_{mod}$  and  $\kappa$ , respectively (Fig. 2B), reflecting the natural Pb background noise of the Medjerda River and the surrounding Pliocene rocks (5.3–2.6 Ma) forming the Jebels Kechabta, Touiba, Ennadhour, the *Utica* promontory and the hill of *Castra Corneliana* (29), all of which border the northern part of the delta (Fig. S1).

**Tunisian mine tailings as the source of Pb pollution**

Just under half of the samples (69 of 147) have Pb concentrations elevated well above background level, which must derive from anthropogenic pollution. Three lines of evidence point to the source of the Pb pollution in the Medjerda delta being mine tailings in the river’s watershed.

(i) The 69 Pb-polluted sediments differ from the unpolluted samples in both their Pb content and Pb isotopic composition. Lead concentrations and  $EF_{Pb}$  range between 10 and 210 ppm (mean = 59.1 ppm) and 2.13 and 20.9 (mean = 6.9), respectively (Fig. 1 and Dataset S1). According to the  $^{14}\text{C}$  age-depth model

reconstructed for the UKC1 reference core (Fig. 1 and Dataset S2), the first Pb pollution event started in the middle of the 4<sup>th</sup> c. BC, when *Utica* was under the hegemony of Carthage. This anthropogenic Pb enrichment occurs in the UKC1 reference core between 5.8 and 8.2 m core depth and shows up as a decrease in <sup>206</sup>Pb/<sup>204</sup>Pb and <sup>208</sup>Pb/<sup>204</sup>Pb isotopic ratios (respectively <18.8 and <38.9) and an increase in <sup>207</sup>Pb/<sup>206</sup>Pb and <sup>208</sup>Pb/<sup>206</sup>Pb ratios (respectively >0.835 and >2.07) (Fig. 1 and Dataset S1). More generally for the Medjerda delta as a whole, a lead pollution event caused by an anthropogenic source is attested by a surge of exotic lead observed in leachates with values above 0.0532 and 2.070 for <sup>204</sup>Pb/<sup>206</sup>Pb and <sup>208</sup>Pb/<sup>206</sup>Pb, respectively (Fig. 2A). This change in the local Pb isotopic composition also affects the residual fraction of Pb since its <sup>208</sup>Pb/<sup>206</sup>Pb increases until overlapping the anthropogenic Pb of the UKC1 core between 5.8 and 8.2 m core depth (highlighted by the gray band and red stars in Fig. 1A). This co-evolution of leachate-residue pairs is seen in the downcore behavior of the isotopic contrast between the residue and the leachate ( $\Delta$ Pb isotope ratios) in the UKC1 core (Fig. 1A). Values of  $\Delta$ Pb isotope ratios in the anthropogenic Pb level of the UKC1 core (between 5.8 and 8.2 m core depth) are  $\sim 0$ , meaning that the Pb isotopic compositions within these physico-chemical Pb fractions are the same. Usually the opposite occurs during a pollution phase (11, 26). This concordance between residues and leachates is also observed for the geochemical parameters  $T_{\text{mod}}$  and  $\kappa$  (as well as  $\mu$ , the <sup>238</sup>U/<sup>204</sup>Pb ratio) which, at the anthropogenic Pb level of the UKC1 core (between 5.8 and 8.2 m core depth), display similar increasing values of, respectively, > 30 Ma and 3.91 (Fig. S2). Such co-evolution of leachate-residue pairs (Figs. 1, 2B and S2) implies that the labile and crustal lead have a common anthropogenic source, which in turn must have a sedimentary origin (the residual fraction cannot come from somewhere else). We therefore suggest that the only conceivable anthropogenic Pb source is that of early mining activity involving Pb ores and the release of mine tailings into the Medjerda watershed.

(ii) The idea of a mining source for the Pb-contaminated sediments in the *Utica* cores is supported by the alignment of the core samples in a plot of  $T_{\text{mod}}$  vs.  $\kappa$  along two mixing lines with a natural end-member  $\alpha$  and an anthropogenic end-member  $\beta$  (Fig. 2B). The Hercynian Pb model ages of the  $\beta$  component ( $T_{\text{mod}} \sim 140\text{--}240$  Ma) split into two local subcomponents  $\beta'$  and  $\beta''$ , which correspond to some modern Tunisian mining districts (Fig. 2B). The  $\alpha$  end-member has an epicenter on a Pb model age of  $\sim 3.8$  Ma (Fig. 2B), which closely matches that of the Pliocene rocks (5.3–2.6 Ma) surrounding the northern part of the Medjerda delta. The mixing lines  $\alpha\text{--}\beta'$  and  $\alpha\text{--}\beta''$  are distinguished according to the periods to which the Pb-contaminated sediments refer, and the metalliferous sources from which they are derived (Fig. 2B). It is not surprising to find a significant contrast between the Pb isotopic signatures of certain metalliferous deposits within the Medjerda catchment basin and its deltaic sediments given the specific mineralogical history of the metal ores forming the mining districts which is distinct from that of the host rocks (26). Similar cases have been documented, for example, in Bulgaria (30), Iran (31) and Mexico (32). For Tunisia, the isotopic contrast is all the more pronounced because its metallic resources, almost exclusively galena and sphalerite (33, 34), are hosted by tectonically, lithologically and stratigraphically widely diverse environments which, moreover, cover geological periods from the Triassic to the Upper Miocene (5–253 Ma) (8, 35, 35).

(iii) In addition to the co-evolution of Pb leachate-residue pairs in the *Utica* core sediments (Figs. 1 and S2) and the Pb isotope mixing lines pointing towards Tunisian ores (Fig. 2), another indicator of mine tailings being the only source of Pb pollution in the *Utica* sediments comes from elemental geochemistry (Dataset S3). In Fig. S3, factor analysis of the bulk sediment element

abundances identifies Pb as clustered tightly with Ag ( $r \sim 0.92$ ), P ( $r \sim 0.94$ ), Cu ( $r \sim 0.79$ ) and Zn ( $r \sim 0.62$ ), with a large loading on the first factor indicative, respectively, of a common source for all these elements, and of their predominance in the sedimentary deposits of the Medjerda's delta. According to the correlogram of the 29 elements analyzed, the highest positive Pb correlations are found with these elements (Fig. S4). Recently, such factor analysis has contributed to identifying lead pipes as the Pb pollution source of the ancient harbor sediments of Naples because Pb was clearly separated from other metal trace elements, thus indicating the presence of a pure Pb source (10). Here the factor analysis shows the opposite, with an impure anthropogenic Pb source containing Pb, Ag, P, Cu and Zn. The most common Ag ore is galena, a lead sulfide. Most silver mines, therefore, are also lead mines. Galena is today exploited at several sites in Tunisia (37, 38), with an average Ag content ranging from 200 to 500 g/t of galena (36), values sufficiently high to suggest its exploitation for silver as well as lead during ancient times (9, 36, 37). In Spain, for example, mining archaeology has shown that Roman lead-silver mines have an Ag content ranging from 50 to 8000 g/t of galena (36). In the Sierra Morena in Spain, a hotspot of ancient lead-silver mining, Domergue (37) concluded that the Ag content in galena is generally between 300 and 500 g/ton. Copper can occur either as traces in the galena or as a result of chalcopryrite extraction. Nevertheless, this last hypothesis is unlikely in the case of Tunisian trace element systematics since ore deposits hosting this type of Cu mineral are very rare in Tunisia (34). Zinc deposits, in contrast, largely associated with those of Pb because Pb and Zn are very similar geochemically, are widespread in Tunisia (10, 34, 35) and consist of blende and smithsonite ores (34). Finally, phosphorus is not known to be associated with galena, implying that its grouping with Pb may attest the simultaneous exploitation of Tunisian phosphate deposits (34).

Based on the three geochemical arguments above (the co-evolution of Pb leachate-residue pairs, Pb isotopic mixing lines with Tunisian ores as the anthropogenic end-member, and clustered metal trace element systematics), this study has made it possible for the first time to establish direct measurable evidence of ancient mining activity in the Medjerda catchment basin.

### Tracing the Tunisian mining sources

Rather than using the classical elliptical fields technique to identify the mining districts responsible for environmental contamination (e.g. 21), we here favor the use of mapped-out Pb isotope data from compiled Pb isotope databases. In addition to providing full transparency on the data used to build these databases (27), this technique has demonstrated its usefulness in various contexts, including, but not limited to, archaeometry (27) and environmental sciences (10). Figure 3 is a map of the compiled Pb isotope data ( $n = 135$ ) for Tunisian metal ores (mainly galena) (see SI Text for references) represented in terms of  $T_{\text{mod}}$  (Fig. 3A) and  $\kappa$  (Fig. 3B). This particular geographical display of the data shows at a glance that the isotopically suspected Pb-bearing districts are part of the Medjerda catchment area, supporting the argument above that the co-evolution of Pb leachate-residue pairs requires a mining source of pollution within the Medjerda watershed.

The first generation of strongly Pb-contaminated sediments dating back to the Punic period plots along the mixing line  $\alpha\text{--}\beta'$  (Fig. 2B), where the anthropogenic subcomponent  $\beta'$  refers to the mines of Djebba and Kebbouch (Fig. 3). Both have Pb isotopic compositions consistent with that of sub-component  $\beta'$  ( $T_{\text{mod}} \geq 140$  Ma and  $\kappa \geq 4$ ) (Fig. 3) and are located inside the Medjerda watershed, unlike the mines of Hamra or Sekarna (Fig. 3). It has long been suspected that the Djebba mines, in the lower Medjerda plain, were exploited since Punic times (15, 16), while those of Kebbouch are thought to have been exploited only in modern times (34). It is not possible to know at which site the

Pb and Ag ores were mined, or whether production took place simultaneously at both sites.

The second cluster of strongly Pb-contaminated sediments dates to the Punic and Roman periods and plots along the mixing line  $\alpha$ - $\beta''$  (Fig. 2B), where the anthropogenic subcomponent  $\beta''$  refers to the Pb-Ag mines of Slata and Bou Jaber ( $T_{\text{mod}} \geq 200$  Ma and  $\kappa \geq 4$ ) (Fig. 3). The Pb isotopic signatures of Hamra, Chaambi-Agab and Sekarna also match that of the subcomponent  $\beta''$  (Fig. 2), but these sites are not within the Medjerda catchment (Fig. 3). Like the mines of Djebba, the Bou Jaber mines are suspected to have been exploited since antiquity (8, 17), while the Slata deposits are believed to have been worked only since medieval times (8, 33).

#### A historical chronology of Tunisian Pb pollution

While the periods defined below correspond to what history has described as phases of instability, the reality was more complex. Each period was marked by both a series of geopolitical conflicts and quieter times. Instability is thus defined here as an increase in the frequency of conflicts compared to preceding or succeeding periods.

##### *Phase 1 (340–280 BC): mining during the time of the Greco-Punic Wars (480–307 BC)*

Although Carthage was founded by Phoenician settlers at the beginning of the 1<sup>st</sup> millennium BC, the first phase of Pb contamination in the deltaic sediments of the Medjerda River did not occur until the second half of the 4<sup>th</sup> c. BC. At this time a slight increase in Pb concentrations is recorded ( $EF_{\text{Pb}} = 3$ –11) in the cores UKC1 (Figs. 1, S3 and S2) and UTC12 (Dataset S1), as well as an aging of the Pb model ages centered around 40 Ma (note in Fig. 2B, the three Punic samples plotting outside the Pb isotopic composition of the natural background). This signal probably represents the first mining activity in Tunisia, which seems to have developed during the extension of Carthaginian control over its hinterland from the 5<sup>th</sup> c. BC, once the First Greco-Punic War ended (480 BC) (38). According to the <sup>14</sup>C age-depth model, this early phase of mining in the Medjerda catchment extends from c. 340 to 280 BC. Particularly striking is that the onset of this early pollution phase coincides with the first minting of Punic coins at Carthage during the middle of the 4<sup>th</sup> c. BC (39): after that the Carthaginian economy became increasingly monetized (40). The minting of silver coinage intensified during the conflict against Syracuse (317–289 BC), which has led some scholars to consider this war as the trigger for monetary activity at Carthage (40, 41).

##### *Phase 2 (275–180 BC): increasing mining activity in the first two Punic Wars (264–201 BC) and the inter-war period (241–218 BC)*

The anthropogenic excess Pb trapped in deltaic sediments rises sharply from 275 BC (Figs. 1 and S3) with  $EF_{\text{Pb}}$  values of up to 20 (Fig. 1 and Dataset S1). Pb isotopes indicate that between 275 and 180 BC (sediments dated to the Punic period that plot along the mixing line  $\alpha$ - $\beta'$  in Fig. 2) the mining districts of Djebba and/or Kebbouch (Figs. 2 and 3) were mined with tailings discharged into the Medjerda catchment, contributing to increasing Pb levels in the local sediments. This second Tunisian mining phase took place during the first two Punic Wars (264–201 BC). As with the first mining phase (340–280 BC), activity during this period seems to have been driven by a military context requiring significant financial and monetary resources (1, 2). Numismatic studies show that from the beginning of this conflict Carthage increased its minting of silver coins (39, 40) to pay its mercenaries (42). It can now be assumed that part of the lead-silver ores contained in the coins minted at Carthage was extracted from the mines of the Medjerda watershed.

Moreover, because the Pb and Ag mines of Djebba and/or Kebbouch were exploited for almost a century (275–180 BC), it can be assumed that these mining districts also contributed to funding the penalties imposed by Rome on Carthage during this conflict: 3,200 talents of silver (96 tons) (43) at the end of the

First War, and 12,500 talents (375 tons) after the Second War (44). Indeed, while it has generally been thought that the penalty of the First Punic War (264–241 BC) was paid in silver from the Iberian peninsula (1), we now suspect that the Tunisian mines also contributed to this effort since Carthage successively lost its traditional silver sources during the first two Punic Wars: Sardinia and Sicily in 241 BC, and Spain in 201 BC.

##### *Phase 3 (180–95 BC): sustained mining activity until the end of the Punic Wars*

The recorded levels of Pb pollution between 180–95 BC are still high as shown by  $EF_{\text{Pb}}$  values ranging between 10 and 20 (Fig. 1 and Dataset S1). The Pb isotope ratios are consistent with this observation since <sup>206</sup>Pb/<sup>204</sup>Pb displays a decrease in values to ~18.6 (natural <sup>206</sup>Pb/<sup>204</sup>Pb > 18.8), while <sup>207</sup>Pb/<sup>206</sup>Pb and <sup>208</sup>Pb/<sup>206</sup>Pb are, respectively, > 0.835 and 2.07 (Fig. 1). This period differs from the second period of mining activity because of a change in the metalsources exploited by the Carthaginians, and later by the Romans, as indicated by the alignment of the corresponding samples along the second mixing line  $\alpha$ - $\beta''$  (Fig. 2). Lead and silver mining was now evidently carried out in the mines of Bou Jaber and/or Slata (subcomponent  $\beta''$ ) from the beginning of the 2<sup>nd</sup> c. BC onwards (Fig. 3).

Once again, this major event took place in a particular historical context: the interval between the Second and Third Punic Wars (201–149 BC). During this period, Carthage suffered a double financial pressure: it no longer had any metal resources in Spain, and was forced to pay war compensations in Rome about four times the level of those after the First Punic War. There was thus a considerable imbalance between Carthage's monetary needs and the metalresources available to it. We can surmise that this strong demand for mineral resources would have pushed the Carthaginians towards mining districts with higher potential and located in the territory it still controlled (modern Tunisia). Paradoxically, this period between the two last Punic Wars is considered to be a period of renewed prosperity in Carthage, whose traces can be seen in the city's urban planning (45), and apparently reflecting a revival of commercial activities based on the export of ceramics, cattle, and, potentially, metals (1). Our new results suggest that the Carthaginians were exceptionally economically resilient during this period, owing to their exploitation of mining regions in Carthage's hinterland.

Figure 2 shows that the sediments deposited after the Third Punic War (149–146 BC) remain affected by Pb excesses, but to a lesser extent (samples are closer to the natural component  $\alpha$ ), resulting from mining Pb-Ag ores from Slata and/or Bou Jaber (Fig. 3), from then on under Roman control. It is difficult to determine whether the Romans' desire to appropriate the Carthaginian mines was one of the causes of the Third Punic War, but their appropriation was certainly a consequence of it. After going to Carthage in 157 BC, Cato was struck by the prosperity of Africa and later became a fervent advocate for the destruction of Carthage (44). Carthage had settled its war debt to Rome, which may have been tempted permanently to eliminate its old rival, monopolizing its metal resources at the same time. Roman history later shows that once a territory was annexed its metal resources were swiftly exploited for the benefit of the imperial power (6); the Carthaginian hinterland would have undergone this same dynamic, as Carthaginian territory had already done in Sicily and Spain.

##### *Phase 4 (95 BC–AD 800): declining mining activity*

Figure 2 shows that the samples from the Republican period until the Early Middle Ages are still aligned along the mixing line  $\alpha$ - $\beta''$ , but with a greater proportion of the natural component  $\alpha$ . Consequently, it appears that at the beginning of the 1<sup>st</sup> c. BC, mining activity in the Medjerda catchment in the mining districts of Bou Jaber and/or Slata continued, but much less intensively. This slowdown in metal exploitation during the Roman period

in Tunisia must be seen in the light of Roman territorial expansion northward into northern Spain, Gaul, Germany and Britain, where Rome gained access to new metal resources with potential higher than that of the Carthaginian mines (6, 7).

Mining activity continued through the early medieval period until the beginning of the 9<sup>th</sup> c. The latest Pb-polluted levels recorded during the medieval period are in the peat layers of cores UTC1, 2 and 12 (Fig. 2), which display  $EF_{Pb}$  and  $T_{mod}$  values ranging between 2 and 8 and 46 and 97, respectively (Dataset S1).

### Perspectives

From this research emerge multi-disciplinary perspectives relating to history, archaeology, and geomorphology. From a historical point of view, the occurrence of a first phase of anthropogenic Pb pollution not before the middle of the 4<sup>th</sup> century BC excludes the idea that it was the attraction of the mineral resources of the Carthaginian hinterland that led to the original settlement of the Phoenicians at Carthage and *Utica* at the beginning of the 1<sup>st</sup> millennium BC. From an archaeological point of view, this study reveals the potential of the Tunisian mining deposits, and those of North Africa more generally, for archaeological mining studies. Finally, from a geomorphological point of view, the considerable mine tailings released during the ancient periods of mining activity documented here should be regarded as an important factor alongside climate and land use for the evolution of deltas. The current environmental effects of Tunisia's mining legacy are a matter of concern as remobilization of Roman mining tailings could be triggered by fluvial erosion processes.

1. Le Bohec Y (1995) *Histoire militaire des guerres puniques* (Monaco). Rocher.
2. Hoyos BD ed. (2011) *A Companion to the Punic Wars* (Chichester, UK).
3. Frank T ed. (1933) *An Economic Survey of Ancient Rome* (Johns Hopkins University Press, Baltimore, Maryland, USA).
4. Kay P (2014) *Rome's Economic Revolution* (Oxford University Press, Oxford, UK).
5. McConnell JR, et al. (2018) Lead pollution recorded in Greenland ice indicates European emissions tracked plagues, wars, and imperial expansion during antiquity. *PNAS* 115(22):5726–5731.
6. Domergue C (2008) *Les mines antiques: la production des métaux aux époques grecque et romaine* (Picard, Paris, France).
7. Wilson AI, Friedman H (2010) Mining Database. Available at: <http://oxrep.classics.ox.ac.uk/databases/mines.database/> Webdesign, databasedesign: Miko Flohr, 2010–2018. Content: OXREP, 2005–2018.
8. Skaggs S, Norman N, Garrison E, Coleman D, Bouhler S Local mining or lead importation in the Roman province of Africa Proconsularis? Lead isotope analysis of curse tablets from Roman Carthage, Tunisia. *Journal of Archaeological Science* 39(4):970–983.
9. Tylecote RF (1992) *A History of Metallurgy* (Institute of Materials, London).
10. Delile H, et al. (2016) A lead isotope perspective on urban development in ancient Naples. *PNAS* 113(22):6148–6153.
11. Delile H, et al. (2017) Rome's urban history inferred from Pb-contaminated waters trapped in its ancient harbor basins. *PNAS* 114(38):10059–10064.
12. Longman J, Veres D, Finsinger W, Ersek V (2018) Exceptionally high levels of lead pollution in the Balkans from the Early Bronze Age to the Industrial Revolution. *PNAS* 115(25):5661–5668.
13. Bondi SF (1995) Le commerce, les échanges, l'économie. *La Civilisation Phénicienne et Punique: Manuel de Recherche*, Handbuch der Orientalistik. (Kriings, V., Leiden, Pays-Bas), pp 268–281.
14. Fournel H (1849) *Richesse minière de l'Algérie* (Paris, France).
15. Tissot CJ (1884) *Géographie comparée de la province romaine d'Afrique* ed Reinach S (Paris, France).
16. Berthou H (1922) *L'industrie minière en Tunisie* (Direction générale des travaux publics).
17. Gottis C, Sainfeld P (1952) Les gîtes métallifères tunisiens. Monographies régionales. (La Rapide, Tunis, Tunisie), p 104.
18. Clarke GW (1989) *The Letters of St. Cyprian of Carthage: 4*, Letters 67–82, New York and New Jersey: Mahwah and Newman Press.
19. Uglietti C, Gabrielli P, Cooke CA, Vallelonga P, Thompson LG (2015) Widespread pollution of the South American atmosphere predates the industrial revolution by 240 y. *PNAS* 112(8):2349–2354.
20. Renberg I, Brännvall ML, Bindler R, Emteryd O (2002) Stable lead isotopes and lake sediments—a useful combination for the study of atmospheric lead pollution history. *Sci Total Environ* 292(1–2):45–54.
21. Véron AJ, et al. (2013) A 6000-year geochemical record of human activities from Alexandria (Egypt). *Quaternary Science Reviews* 81:138–147.
22. Delile H, et al. (2015b) The geoarchaeology of Utica (Tunisia): the palaeo-geography of the Mejerda delta and hypotheses concerning the location of the ancient harbour. *Geoarchaeology* 4:291–306.
23. Pleuger E, et al. (In Press) Exploration of the maritime façade of Utica: the potential location of the Phoenician and Roman harbours. *Quaternary International*.

## Materials and Methods

### Radiocarbon dates and the age-depth model

The 37 radiocarbon dates (22, 23) used in this study are listed in Dataset S2. The measured <sup>14</sup>C (BP) ages were converted into BC–AD dates using the continental and marine curves of Reimer et al. (46). For each core an age-depth model was created using the Clam software (47) to classify the samples according to the broad historical periods covered here (Pre-Punic, Punic, Republican, Early Roman Empire, Late Roman Empire, Early Middle Ages and Late Middle Ages).

### Major and trace element analysis

Major and trace element analyses were carried out on core UKC1 (10 m depth) with a sample resolution of one sample approximately every 12 cm (86 samples analyzed in total) (Dataset S3). The analytical details are those of Delile et al. (10, 11), which are briefly summarized in the SI Text.

### Pb isotopic analysis

Lead isotopic compositions were measured on 146 leachates and 79 residual fractions, the latter of which were from the same sample digestions as the leachates. The samples were distributed among the eight cores of this study as follows (Dataset S1): UKC1, 86 leachates and 30 residues; UTC2, 1 leachate; UTC1, 3 leachates; UTC10, 5 leachates and 5 residues; UTC12, 11 leachates and 7 residues; UCN2, 18 leachates and 15 residues; KAL1, 17 leachates and 17 residues; and UCN1, 5 leachates and 5 residues. The analytical details are those of Delile et al. (10, 11) with the main steps summarized in the SI Text.

**ACKNOWLEDGMENTS.** We thank the Roman Mediterranean Ports program (ERC grant agreement no 339123), which funded this study, and the National Heritage Institute of Tunisia (INP) for the authorization to access the archaeological area of Utica in which some of the cores were taken. The ARTEMIS-SHS program and radiocarbon laboratory of Lyon performed the radiocarbon dating. We are grateful to Philippe Telouk for ensuring that the mass spectrometers were always working well.

24. McLennan SM (2001) Relationships between the trace element composition of sedimentary rocks and upper continental crust. *Geochemistry, Geophysics, Geosystems* 2(4):24.
25. Cheng H, Hu Y (2010) Lead (Pb) isotopic fingerprinting and its applications in lead pollution studies in China: a review. *Environ Pollut* 158(5):1134–1146.
26. Bird G (2011) Provenancing anthropogenic Pb within the fluvial environment: Developments and challenges in the use of Pb isotopes. *Environment International* 37(4):802–819.
27. Albarède F, Desauty A-M, Blichert-Toft J (2012) A geological perspective on the use of Pb isotopes in Archaeometry. *Archaeometry* 54(5):853–867.
28. Blichert-Toft J, et al. (2016) Large-scale tectonic cycles in Europe revealed by distinct Pb isotope provinces. *Geochemistry Geophysics Geosystems* 17(10):3854–3864.
29. Burolet PF, Dumon E (1952) Carte géologique de la Tunisie, Porto-Farina.
30. Bird G, et al. (2010) Quantifying sediment-associated metal dispersal using Pb isotopes: application of binary and multivariate mixing models at the catchment-scale. *Environ Pollut* 158(6):2158–2169.
31. Shafiei B (2010) Lead isotope signatures of the igneous rocks and porphyry copper deposits from the Kerman Cenozoic magmatic arc (SE Iran), and their magmatic-metallogenetic implications. *Ore Geology Reviews* 38(1):27–36.
32. Cameron KL, et al. (1992) Contrasting styles of Pre-Cenozoic and Mid-Tertiary crustal evolution in northern Mexico: Evidence from deep crustal xenoliths from La olivina. *Journal of Geophysical Research* 97:17353–17376.
33. Sainfeld P (1956) The lead-zinc-bearing deposits of Tunisia. *Economic Geology* 51(2):150–177.
34. Djellouli A, Ilavsky J, Nicolini P (1968) Carte des gîtes minéraux de la Tunisie.
35. Bouhler S (2007) Les ressources en plomb, zinc, fer, argent, cuivre, or, barytine, fluorine et césistite de la Tunisie : un bilan de 117 ans de recherches, d'explorations et d'exploitations (Hammamet), p 2.
36. Domergue C (1987) *Catalogue des mines et des fonderies antiques de la péninsule Ibérique* (Casa de Velázquez, Madrid).
37. Domergue C (1990) *Les mines de la péninsule Ibérique dans l'antiquité romaine* (École française de Rome, Rome, Italie).
38. Decret F (1992) *Carthage ou l'Empire de la mer* (Seuil, Paris, France).
39. Jenkins GK, Lewis RWB (1963) *Carthaginian gold and electrum coins* (Royal numismatic society, London).
40. Visonà P (1998) Carthaginian coinage in perspective. *American Journal of Numismatics* 10:1–27.
41. Mildner L (1989) Punic Coinage on the Eve of the First War against Rome. A Reconsideration. *Punic Wars*, Studia Phoenicia, 10. (Devijver H., Lipinski E., Leuven), pp 5–14.
42. Harl KW (1996) *Coinage in the Roman economy, 300 B.C. to A.D. 700* (The John Hopkins university press, Baltimore, London).
43. Polybius, *The Histories* (William Heinemann, London), The Loeb classical library.
44. White H ed (1913) *Appian, Roman History* vol. 1, Books 1 - 8.1 (Harvard University Press, Cambridge), Loeb Classical Library 1, at Book 8.X.69.
45. Lancel S ed. (1979) *Byrsa* (École française de Rome, Rome).
46. Reimer P, et al. (2013) IntCal13 and Marine13 Radiocarbon Age Calibration Curves 0–50,000 Years cal BP. *Radiocarbon* 55(4):1869–1887.
47. Blaauw M (2010) Methods and code for 'classical' age-modelling of radiocarbon sequences. *Quaternary Geochronology* 5(5):512–518.
48. Anzidei M, et al. (2011) New insights on the relative sea level change during Holocene along

817	the coasts of Tunisia and western Libya from archaeological and geomorphological markers.	carried out the field work; H.D. and J.B.-T. produced the data; H.D., E.P., J.B.-T., N.F., E.F.,	885
818	<i>Quaternary International</i> 232(1-2):5-12.	and A.I.W. analyzed and interpreted the data; H.D., J.B.-T., E.F., and A.I.W. wrote the paper.	886
819	<b>Author Contributions</b>		887
820	H.D. and J.-P.G. designed the project; H.D., E.P., J.-P.G., A.G., H.A., I.B.J., E.F., and A.I.W.		888
821			889
822			890
823			891
824			892
825			893
826			894
827			895
828			896
829			897
830			898
831			899
832			900
833			901
834			902
835			903
836			904
837			905
838			906
839			907
840			908
841			909
842			910
843			911
844			912
845			913
846			914
847			915
848			916
849			917
850			918
851			919
852			920
853			921
854			922
855			923
856			924
857			925
858			926
859			927
860			928
861			929
862			930
863			931
864			932
865			933
866			934
867			935
868			936
869			937
870			938
871			939
872			940
873			941
874			942
875			943
876			944
877			945
878			946
879			947
880			948
881			949
882			950
883			951
884			952

# Submission PDF