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1 **The contribution of geochemistry to ancient harbor geoarcheology: The**
2 **example of Ostia Antica**

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12 **Abstract**

13 Once trapped in ancient harbor basins, sediments form environmental archives that have been
14 widely studied by geoarcheologists in recent decades, especially to help reconstruct fluvio-
15 marine landscapes of the last millennia. In some cases, classic environmental markers cannot
16 be used for this purpose either because of their scarcity in the sedimentary deposits, or
17 because analytical costs limit the resolution that can be achieved. In order to remedy these
18 shortcomings, and to complement the more commonly used proxies, elemental and isotopic
19 geochemistry has been added to the geoarcheological toolkit. Here we show how to “read” the
20 evolution of the paleo-environmental dynamics in the water column of Ostia Antica (Rome’s
21 first maritime harbor) using the geochemical and isotopic record of a 3000-year-old sediment
22 core drilled in the ancient harbor basin. A comparison of the results obtained from Ostia
23 Antica with those of other ancient Mediterranean harbors reveals the nature of the main
24 environmental processes operating during the formation of sedimentary deposits in harbor

25 basins. From this comparative approach, it appears that the respective weight of each control
26 factor is dependent on the coastal geomorphological context of the sites where the harbors
27 were established. Since the discovery of the harbor of Ostia Antica in 2014, this method has
28 provided the means, for the first time, to identify two distinct harbor basin regimes; an initial
29 marine-dominated regime from the middle of the 4th c. BC to ~ the 3rd c. BC, and a later
30 freshwater-dominated regime up to the 2nd c. BC. More generally, we observe the effects of
31 the dynamics of the deltaic progradation of the Tiber, which very early on was subject to a
32 hydro-climatic component, on the processes of alluviation of the harbor basin. Additionally,
33 and also for the first time in harbor geoarcheology, Pb isotope compositions measured
34 specifically on uncontaminated sediments demonstrate their utility for both identifying the
35 geological sources of the sediments of the Tiber delta and discriminating finer from larger
36 particles. The present study further provides an opportunity to test the validity of two
37 hypotheses recently put forward: (1) that a series of three tsunamis is recorded in the harbor
38 silts, and (2) that an initial lagoon-type harbor was constructed at Ostia Antica, which later
39 evolved into a fluvial harbor. Neither of these hypotheses are supported by the present data.

40

41 **Keywords:** Rome, Ostia Antica, ancient harbor basins, geoarcheology, sedimentology,
42 geochemistry, Pb isotopes, Tiber river

43

44 **1. Introduction**

45 When harbor geoarcheology emerged as a discipline at the beginning of the 1990s
46 following the excavations of the ancient harbors of *Caesarea* (Israel) (Reinhardt et al., 1994,
47 1998; Reinhardt and Raban, 1999) and Marseille (France) (Hesnard, 1994; Morhange, 1994;
48 Morhange et al., 2001, 2003), the analytical and investigative methods used were based on
49 those well known from other domains within the earth sciences, such as micropaleontology

50 (molluscs, ostracods, foraminifera, diatoms, pollen) and sedimentology (texture,
51 granulometry, exoscopy) (Goiran and Morhange, 2003; Marriner and Morhange, 2007;
52 Bravard, 2009; Cubizolle, 2009). Two decades later, while these well-tested methods continue
53 to be used in the field, elemental and isotopic geochemistry has been added successfully to the
54 geoarcheology toolkit, as we show below.

55 Although geochemistry began to be used in harbor geoarcheology in the mid-2000s to
56 determine human impact based on findings of ancient metal traces (Le Roux et al., 2003,
57 2005; Véron et al., 2006, 2013, 2018; Marriner and Morhange, 2007; Stanley et al., 2007;
58 Delile et al., 2014b, 2015, 2016b, 2017), it had not so far been used in paleo-environmental
59 studies. It is only very recently that research teams, primarily from Germany, have used
60 elemental geochemistry to improve on paleo-environmental reconstructions in archeological
61 contexts. Important examples of such work carried out on ancient harbors are the studies of
62 Oiniadai (Vött, 2007), Olympia (Vött et al., 2011), Palairos-Pogonia (Vött et al., 2011),
63 Corinth (Hadler et al., 2013a), Kyllini (Hadler et al., 2015b), and Alkinoos (Finkler et al., 2018)
64 in Greece; Ephesus (Stock et al., 2013, 2014, 2016; Delile, 2014; Delile et al., 2015) and Elaia
65 (Shumilovskikh et al., 2016) in Turkey; *Portus* and Naples in Italy (Delile et al., 2014a,
66 2016a); and Magdala (Rossi et al., 2015) in Israel. This recent expansion of geochemistry into
67 geoarcheological research is a direct result of the widespread use of micro-XRF core scanners
68 during the last decade. This technology can rapidly, non-destructively, and automatically scan
69 sediment cores to produce elemental profiles at high resolution, which contribute greatly to
70 the reconstruction of paleo-environments (Croudace and Rothwell, 2015). However, to take
71 full advantage of this technique, it is indispensable to have prior knowledge of the
72 geochemical properties of the sediments in question in order to avoid misinterpretations. One
73 such common mistake consists in using certain elemental ratios capable of tracing the
74 sedimentological (e.g. Zr/Rb and Zr/Ti applied as particle size proxies) or environmental (e.g.

75 Sr/Al, Mg/Ca or Ca/Al, and Si applied as biological production proxies or Mn/Fe, U, and Mo
76 applied as water ventilation proxies) parameters without taking into account the geological,
77 hydrological, and climate contexts of the sites under study, each of which can significantly
78 modify the meaning of these ratios. Initially, the use of geochemistry for reconstructing
79 environmental processes was applied to (i) marine sediments to decipher paleo-productivity,
80 deep water ventilation, and paleo-temperatures, and (ii) lacustrine sediments to track
81 atmospheric pollution, human impact, paleo-climatic changes, aeolian input, paleo-floods,
82 catchment weathering, and source rock compositions (Boyle, 2002; Croudace and Rothwell,
83 2015).

84 Here, we discuss the types of environmental and anthropogenic processes that are most
85 likely to benefit from the use of high-resolution geochemistry applied to the geoarcheological
86 study of ancient harbors. To that end, we identify the geochemical fingerprints of the main
87 environmental and anthropogenic processes that took place in the ancient harbor basin of
88 Ostia Antica and compare them with:

- 89 (i) the “classic” environmental archives documented so far by geochemistry (i.e.,
90 lacustrine and marine sediments);
- 91 (ii) the “traditional” proxies used in harbor geoarcheology (see above), especially those
92 applied to Ostia Antica’s ancient harbor basin, which are well documented from a paleo-
93 environmental perspective (Goiran et al., 2012, 2014, 2017; Hadler et al., 2015a; Sadori et
94 al., 2016; Wunderlich et al., 2018);
- 95 (iii) those identified at *Portus* (Delile et al., 2014a for the Trajanic basin and this study for
96 the Claudius basin), Ephesus (Delile et al., 2015), and Naples (Delile et al., 2016a).

97 Although micropaleontological (ostracods, foraminifera, pollen) and sedimentological
98 (texture, granulometry) studies have been carried out on the harbor deposits of Ostia Antica,
99 the dynamics and paleo-environmental changes in its water column remain poorly understood.

100 Improving this understanding is all the more important since Ostia Antica had a river harbor
101 located a short distance from the mouth of the Tiber River, which was to subject it to both
102 marine and river influences (Goiran et al., 2012, 2014, 2017; Salomon et al., 2016). As a
103 result, in the present work, we focus in particular on the interaction between fluvial and
104 marine impacts on the harbor water column.

105 2. Study area

106 2.1. Archeological and historical background

107 According to Roman historians, Ostia Antica was founded during the reign of Ancus
108 Marcius in the 7th c. BC. However, the *castrum* (building, or plot of land, used as a fortified
109 military camp) did not appear before the 4th or beginning of the 3rd c. BC (Coarelli, 1988;
110 Martin, 1996; Zevi, 2002; Brandt, 2002). The absence of evidence for the earliest periods has
111 led to hypotheses about the existence of a paleo-Ostia, the location of which remains
112 unknown and would correspond to the city founded by Ancus Marcius (Coarelli 1988; Martin
113 1996; Zevi 2002; Brandt 2002). Ancient texts reveal that the harbor of Ostia Antica became a
114 commercial and military port during the Punic wars (Livy, 45, 22) and played a vital role in
115 providing Rome with food supplies (especially olive oil from Spain and grain from the North
116 African provinces) during the 1st c. BC (Le Gall, 1953; Zevi, 2001b). Nevertheless, at this
117 time, Puteoli (Pozzuoli) on the Bay of Naples remained the principal maritime port of Rome
118 for trade (Balland, 1965). It was considered to be one of the busiest centers of trade in the
119 Roman Empire, especially for metal ingots, for which a route from the Cartagena/Mazarron
120 and Rio Tinto mines in Spain to Puteoli and Rome was heavily used (Domergue and Rico,
121 2014; Delile et al., 2016b). The large distance separating the ports of Puteoli and Ostia Antica,
122 as well as a problematic massive siltation of Ostia's harbor basin caused by a succession of
123 floods discharging sediments at the mouth of the Tiber (Strabo, 5, 3, 5), combined with the
124 need to accommodate an increasing number of merchant and military ships arriving from the

125 Roman provinces, were the main factors that led to the foundation of the new *Portus* harbor
126 complex 3 km north of Ostia Antica in the middle of the 1st c. AD (Keay et al., 2005; Goiran
127 et al., 2014) (Fig.1A).

128 2.2. *Geography and geology*

129 The Tiber delta constitutes the outlet for the water and sediment discharges of a river
130 405 km long which drains a watershed surface area of 17,375 km², comprising young
131 sediments from the Apennines and modern volcanic deposits in Latium. The annual
132 deposition of sediment in the Tyrrhenian Sea, estimated at 7.2 million tons per year (Iadanza
133 and Napolitani, 2006), has resulted in the construction of a wave-dominated delta 150 km² in
134 extent. The Tiber delta can be divided into (i) the eastern inner deltaic plain, related to the
135 paleo-lagoons of Maccarese in the north and Ostia in the south; (ii) the western outer deltaic
136 plain, where the landscape is composed of accreting dunes to which must be added the
137 submerged area; (iii) the deltaic front (mainly composed of sand and silt); and (iv) the
138 prodelta (mainly composed of mud and clay) (Salomon, 2013; Salomon et al., 2017).

139 The geomorphological evolution of the Tiber delta during the Holocene period is well
140 documented (Bellotti et al. 1994, 1995, 2007, 2011; Giraudi, 2004; Milli et al., 2013;
141 Salomon, 2013; Salomon et al., 2018). During the periods of the Republic and the Empire, the
142 position of the Tiber mouth likely was located close to the Boacciana Tower (Tomassetti
143 1897) (Fig.1B). Additional information about the coastline comes from the immediate
144 surroundings of *Portus* (Arnoldus Huyzendveld, 2005; Giraudi, 2004; 2009; Salomon, 2013)
145 and from south of the Tiber delta, on the Laurentine shore (Bicket et al., 2009). However,
146 during the Imperial period, the rapid seaward progression of the coastline recorded by the
147 Tiber delta, which could have been more active at the river mouth, is the cause of relatively
148 poor paleo-geographical knowledge of the Tiber mouth and its dynamics during Roman times.
149 This rapid evolution explains why discovering the exact location of the ancient port of Ostia

150 Antica has been so controversial until recently (Goiran et al., 2012). The harbor of Ostia
151 Antica, which has been dated to the period of the Republic, was initially difficult to identify,
152 and its infrastructure (i.e., docks for loading and unloading of goods) could have been located
153 anywhere along the riverbanks in direct connection with the city and its storage facilities
154 (warehouses). But since 2012, a large body of multidisciplinary research combining the work
155 of archeologists, historians, geographers, sedimentologists, geomorphologists, micro-
156 paleontologists, and palynologists has confirmed the location of the river mouth harbor basin
157 of Ostia Antica in the northern part of the city, to the west of the “Palazzo Imperiale”,
158 founded between the 4th and 2nd c. BC (Goiran et al., 2012, 2014, 2017). The stratigraphy of
159 the cores (Goiran et al., 2014) clearly shows the deposit of harbor silts (dated at their base to
160 the middle of the 4th century BC) at a depth of between 5.7 and 9 m below ground level (m
161 *b.g.l.*) (unit B1, Fig. 2) (i.e., between 3.3 and 6.6 m below present sea level (m *b.p.s.l.*) or
162 between 2.5 and 5.8 m below ancient sea level (m *b.a.s.l.*). The sea level between AD 230-
163 450 was defined locally at 0.8 m ± 10 cm below the current biological sea-level by Goiran et
164 al. (2009)). These harbor silts indicate a calm environment with low-energy currents in which
165 the fluvial influence is reflected in the ostracod assemblages (Fig. 2).

166 As shown by Salomon et al. (2016), the stratigraphy of the PO2 core can be considered
167 representative of Ostia’s harbor area as a whole because of (i) its central location in the harbor
168 basin area; (ii) the limited size of this port basin (~ 0.5 ha) in comparison with that of other
169 ancient harbors (for example the Trajanic basin is ~ 39 ha and that of Ephesus in Turkey ~ 20
170 ha); (iii) the consistency of its stratigraphic successions deduced from their spatial correlation
171 with other cores drilled in the harbor (Goiran et al., 2014; Hadler et al., 2015); (iv) the spatial
172 extension of these stratigraphic successions to the entire harbor basin area as inferred from
173 geophysical investigations (Hadler et al., 2015a; Wunderlich et al., 2018); (v) its large number

174 of radiocarbon dates; and (vi) the extraction of the PO2 core by using a rotary mechanical
175 corer that provides cores of a quality superior to those extracted by percussion corers.

176 3. Materials and methods

177 We analyzed at high resolution one (PO2, Fig. 1B) of the two cores drilled by [Goiran et](#)
178 [al. \(2014\)](#) which led to the discovery of the first harbor of Rome at Ostia Antica. This 12
179 meter long core was drilled in a low-lying area without any archeological structural remains
180 near the present-day Tiber River (Fig. 1B).

181 3.1. Radiocarbon dates and the age-depth model

182 The chronostratigraphic framework of the PO2 core is based on 15 radiocarbon dates
183 ([Goiran et al., 2014, 2017; Sadori et al., 2016; Delile et al., 2017](#)) for which the age-depth
184 model was created using the Clam software ([Blaauw, 2010](#)) (Fig. 2). The details of the age-
185 depth model and the ^{14}C dates, as well as the inherent uncertainties associated with both, can
186 be found in [Delile et al. \(2017\)](#), who exercised great caution in the use of these ages by
187 applying them only to broad historical periods (Republic, High and Late Roman Empire, etc.).

188 The singular S-shape of the ^{14}C age-depth model of the PO2 core (Fig. 2) is typical of
189 ancient harbor deposits and due to the dredging practices used at the time. This well-known
190 phenomenon in Mediterranean harbors, well documented at Ostia by [Goiran et al. \(2014,](#)
191 [2017\)](#) and [Salomon et al. \(2016\)](#), is discussed in section 4.4.1.

192 3.2. Analysis of major and trace elements

193 The geochemical analyses were carried out on the entire core with high sample
194 resolution (86 samples analyzed, i.e., one sample approximately every 14 cm). The data are
195 published in [Delile et al. \(2017\)](#) together with the analytical techniques, which are also briefly
196 summarized here. First, the sediments were sieved at 63 μm with a nylon sieve instead of a
197 metal sieve in order to avoid potential contamination. After drying, 100 mg of sieved

198 sediment from each core sample was dissolved at the Ecole Normale Supérieure de Lyon in a
199 clean laboratory under laminar flow hoods. The samples were digested in a 3:1:0.5 mixture of
200 concentrated double-distilled HF, HNO₃, and HClO₄ in Savillex beakers left closed on a
201 hotplate at 120-130 °C for 48 h, then evaporated to dryness. Perchlorates and any remaining
202 fluorides were converted to chlorides by drying down several times with 6 M distilled HCl.
203 The samples in solution in 6 M distilled HCl were all clear, indicating complete sample
204 breakdown. The samples were re-dissolved in 2 ml concentrated distilled HNO₃, from which
205 ~10 percent aliquots were further diluted to 2% HNO₃ and internal standards (10 ppm Sc for
206 ICP-AES and 2 ppb In for Q-ICP-MS) added. Major elements were analyzed by ICP-AES
207 (ICAP 6000) and trace elements by Q-ICP-MS (Agilent 7500 CX). The upper limit of the
208 blank contribution was negligible for major elements and <2% of the sample content for trace
209 elements. The precision on major and trace elements is between 2 and 5%.

210 *3.3. Analysis of Pb isotopes*

211 The method used is described in detail in [Delile \(2014\)](#) and [Delile et al. \(2014b, 2015, 2016b,](#)
212 [2017\)](#) and the main steps summarized in the following. After sieving at 63 µm as described
213 above, representative aliquots of 500 mg of sediment were leached with Suprapur chloroform
214 to separate the labile (or anthropogenic) component of the Pb. A second mild leaching step
215 was performed with dilute double-distilled HBr to further recover the most easily displaceable
216 (presumably anthropogenic) Pb, not all of which could be assumed to have been retrieved by
217 chloroform. The leftover sample residues, constituting the natural “geological” Pb, were
218 discarded. The two leachate fractions were combined and their Pb separated on anion-
219 exchange columns using HBr to elute the sample matrix and distilled 6 M HCl to elute the Pb.
220 The sample residues were not measured in this study because the isotopic composition of the
221 natural Pb background had already been determined in a previous study ([Delile, 2014; Delile](#)
222 [et al., 2014b](#)). The Pb isotope compositions were measured by MC-ICP-MS (Nu Plasma 500

223 HR) at the Ecole Normale Supérieure de Lyon using Tl doping and sample-standard
224 bracketing (Albarède et al., 2004) and the values for NIST 981 of Eisele et al. (2003). The
225 total procedural Pb blank was < 20 pg, far inferior to the multiple µg of Pb leached and
226 separated from each sample. The external reproducibility of the reported Pb isotope ratios as
227 estimated from the repeated (every two samples) runs of NIST 981 are 100-200 ppm (or 0.01-
228 0.02%) for ratios based on 204 ($^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, $^{208}\text{Pb}/^{204}\text{Pb}$) and 50 ppm (or 0.005%)
229 for $^{207}\text{Pb}/^{206}\text{Pb}$, $^{208}\text{Pb}/^{206}\text{Pb}$, and $^{207}\text{Pb}/^{208}\text{Pb}$. The Pb isotope data published in Delile et al.
230 (2017) were used exclusively to focus on Pb anthropogenic excesses, while here we use the
231 same data set to understand the natural, as opposed to the anthropogenic, Pb isotopic signal.

232 **4. Results and discussion**

233 *4.1. Defining the main geochemical fluxes of harbor basin deposits*

234 As mentioned in the introduction, the main objective of this work is to apply multi-
235 elemental analysis to the silico-clastic deposits of the ancient harbor basin of Ostia Antica in
236 order to identify the main inputs and processes involved in their formation. Because the
237 composition and relative weight of different geochemical fluxes reflects the environmental
238 conditions at the time of deposition, we can associate them with environmental variables
239 whose evolution over time enables reconstruction of the paleo-environmental changes that
240 occurred at the scale of the harbor site. For these purposes, the identification of the
241 geochemical fluxes and their associated elementary compositions is a critical aspect requiring
242 great caution.

243 *4.1.1. The three main geochemical inputs and their arbitrarily associated elements*

244 According to Sageman and Lyons (2003), three main inputs are involved in the
245 formation of sedimentary deposits: (i) detrital, (ii) authigenic, and (iii) biogenic materials
246 (Fig. 3).

247 (i) *Detrital material* (Fig. 3A) is derived from erosion of continental crust and volcanic
248 rocks. These erosion products are carried by the hydrological network from their sources at
249 the watershed to the marine environment. The principal elemental components of the particle
250 flux transported by modern fluvial sources are, in order of decreasing abundance, Si, Al,
251 Fe, Ca, K, Mg, Na, Ti, P, Mn, and Ba, according to [Martin and Whitfield, 1983](#). However,
252 inter-comparison of these elements within the PO2 core shows stratigraphic distributions (see
253 panel showing detrital elements in Fig. 3A) that differ from those predicted. This qualitative
254 observation is reproduced quantitatively using the correlogram (Fig. S1) which displays
255 relatively weak levels of correlation between, for example, Al and K ($r = 0.31$), Mg and Fe (r
256 $= 0.06$), and Ti and Ba ($r = 0.2$), even though positive correlations between some elements
257 qualified as detrital, such as Al, Mg, Zr, and Ti, are also observed. These first-order
258 observations demonstrate that it is incautious to use *sensu stricto* certain elements considered
259 to be universal markers of detrital origin. These differences between observation and theory in
260 the chemical composition of the detrital fraction are accounted for by the relative weight of
261 the local specificities that directly affect the factors controlling the composition in question:
262 the mineralogy of the enclosing rocks, the erosional regime which itself depends on regional
263 climate conditions (temperature, precipitation, and runoff), and the reactivity of eroded
264 material during transport ([Sageman and Lyon, 2003](#)).

265 (ii) *Authigenic material* (Fig. 3B) is controlled by the redox state of the system, which
266 depends on the ventilation conditions of bottom waters which in turn determines organic and
267 inorganic reactions by which dissolved constituents are precipitated as minerals, either as
268 metal oxyhydroxides (O_2 enrichment) or metal sulfides (O_2 depletion). The control factors of
269 authigenic material in sediments depend mainly on the water-column mixing rate and the
270 production of organic matter. The redox state usually is tracked by certain elements such as
271 Fe, S, V, Mo, and U ([Sageman and Lyon, 2003](#)), which are assumed to be enriched under

272 reducing conditions and depleted under oxidizing conditions. All of them correlate negatively
273 with Mn because Mn is enriched under oxidizing conditions and depleted under reducing
274 conditions. However, as seen in Fig. 3, the behavior of these elements as a function of
275 stratigraphy does not show such a distribution. This qualitative observation is consistent with
276 the quantitative observation as the correlogram in Fig. S1 shows relatively weak correlations
277 between, for example, Mo and U ($r = 0.07$), V and Mo ($r = 0.2$), and U and S ($r = 0.2$), even if
278 positive correlations are also observed, especially between Mo and Fe ($r=0.55$), Fe and U
279 ($r=0.51$), Fe and V ($r=0.75$), and Fe and S ($r=0.8$) (Fig. S1). None of these elements,
280 therefore, can be used as universal markers of authigenic inputs.

281 (iii) *Biogenic material* (Fig. 3C) reflects the photosynthetic production of organic
282 material and the presence of remains of skeletal material, the latter being composed of calcite
283 (CaCO_3) or silica (SiO_2). Biogenic fluxes in the sedimentary system depend on the
284 concentration and supply of nutrients from surface waters (nitrate, phosphate, and micro-
285 nutrients such as Fe). They are produced by chemical erosion and terrestrial biogenic
286 activity or biological processes in the water column. Usually, the biogenic flux is tracked by
287 Si, Ca, Sr, P, or Loss on Ignition (L.O.I.) (Sagemont and Lyons, 2003; Sánchez Vizcaíno and
288 Cañabate, 1999; Delile et al., 2016a). However, as observed for the detrital and authigenic
289 fluxes, neither the qualitative nor the quantitative observations (Figs. 3C and S1) show clear
290 systematic positive or negative correlations between, for example, Ca and Sr ($r = 0.04$), Ca
291 and P ($r = -0.3$), and Sr and P ($r = 0.3$).

292 This attempt at classification, or grouping, of chemical elements taken individually
293 according to the three principal inputs involved in the formation of sedimentary deposits (Fig.
294 3) has revealed the limits of the approach. First, assigning a given element to a specific
295 category of inputs is not supported by robust statistic correlations, hence calling for caution in
296 automatically using geochemical proxies traditionally considered to be robust tracers for

297 particular inputs. Second, the individual observation of several dozen chemical elements is
298 fastidious work without any real added value. For these reasons, the reading of the underlying
299 causes of geochemical variation and their number is difficult.

300 *4.1.2. A new way to define the main geochemical fluxes of harbor basin deposits*

301 In order to examine in-depth the thousands of data acquired by multi-element analysis (39
302 chemical elements measured) at high resolution (86 samples analyzed in the present study),
303 we use factor analysis in order to convert data into uncorrelated and limited variables, also
304 known as factors. The factors resulting from the statistical treatment of the full data set (39
305 elements for 86 samples) carry a large part (>75%) of the total geochemical variability of the
306 depositional system. These factors in turn correspond to variables that are equivalent to the
307 principal components deduced from the PCA statistical procedure (see Appendix A in [Delile
308 et al., 2014a](#)). Based on the loading values of each factor ([Fig. 4A](#)), the distribution of the
309 chemical elements, which results from positive (element clusters located in the same position
310 relative to the y-axis) and/or negative (element clusters located in the opposite position
311 relative to the y-axis) correlations, enables us to determine the environmental variable
312 expressed behind each factor.

313 Based on this method, we discriminate four main factors controlling the bulk of the
314 geochemical composition of the PO2 core (Fig. 4):

315 (i) *The first factor* (47% of the total variability of the geochemical signal, Fig. 4)
316 comprises elements associated exclusively with a siliciclastic detrital flux (Th, Zr, U, K,
317 REEs, etc.) and is marked with negative F1 values. All these elements are known to be
318 enriched in the Roman Alkaline Province in Italy, including the downstream volcanic rocks of
319 the Tiber River watershed, where they show the highest abundances in floodplain sediments
320 ([De Vos and Tarvainen, 2006](#)). This volcanic province is also known under the name of the

321 Roman Magmatic Province – Latian District (RMP-LD) (Mattei et al., 2010) which regroups
322 the Colli Albani, Vulsini, Cimini, Vicano, Ceriti, and Sabatini volcanoes, all very close to the
323 Tiber delta. The RMP-LD region, which is one of four major volcanic provinces of the
324 Tyrrhenian margins of peninsular Italy, consists of mafic ultrapotassic rocks with a
325 remarkably homogeneous elemental composition characterized by high abundances of Pb, Th,
326 U, Ce, and La and low abundances of Ti, P, Ta, and Nb (Mattei et al., 2010). These
327 geochemical features distinguish all the volcanoes close to the Tiber delta. This is shown in
328 Fig. 4A where elements with affinities for the RMP-LD mafic ultrapotassic rocks show
329 strongly negative values (ranging between -0.7 and -1), whereas elements depleted in the
330 same rocks display significantly lower negative values (ranging between -0.5 and 0).

331 The main characteristic of Factor 1 is that it tracks the sedimentary units with the coarser
332 particles, since the lowest F1 values occur in the sandy units A and B2 (Figs 2, 4B). The
333 correlogram for elementary and granulometry data (Fig. S2) from harbor units only (units B1
334 and B2) shows that the sandy fraction is clustered within these elements (Th, Zr, U, K, REEs,
335 etc.). We conclude from this that Factor 1 reflects siliciclastic terrigenous influence which, in
336 terms of depositional environment, corresponds to the ancient hydrodynamic conditions of the
337 harbor water column.

338 (ii) *The second factor* (13% of the total variability of the geochemical signal, Fig. 4)
339 opposes elements indicative of detrital fluxes (other than the siliciclastic detrital flux of F1)
340 (Ga, Al, Rb, Li, Ti, Mg, etc.) to elements indicative of a carbonate component (Ca). The
341 detrital component (positive F2 values) clearly reflects another type of terrigenous influence,
342 that related to aluminosilicates. Al, Ga, Rb, Li, Mg, and Ti are strongly correlated among
343 themselves (Fig. S1) because they are all constituents of clay minerals (De Vos and
344 Tarvainen, 2006). Figure S2 confirms the predominance (weight) of fine particles in Factor 2
345 because the hierarchical clustering shows good correlations between clays and silts containing

346 these elements. Factor 2 reflects the terrigenous influence of aluminosilicates which,
347 interpreted in terms of an environmental variable, corresponds to the ancient quiet
348 hydrodynamic conditions of the harbor water column.

349 (iii) *The third factor* (10% of the total variability of the geochemical signal, Fig. 4)
350 presents with negative F3 values elements associated with authigenic fluxes (redox
351 conditions), especially those tracking oxygen depletion in the water column (S, Fe, Mo, V).
352 Factor 3 also includes chalcophile elements that precipitate as sulfides under anoxic
353 conditions (Pb, Cu, Co, Sb). This factor also has been recorded in the ancient harbor deposits
354 of Ephesus (Delile et al., 2015).

355 (iv) *The fourth factor* (7% of the total variability of the geochemical signal, Fig. 4)
356 opposes Na to heavy metals such as Sn, Cd, and Pb. The strong influence of Na within the
357 stratigraphy reflects either the salinity of the harbor itself or the invasion of the sediments by
358 the salt wedge. For the Trajanic harbor as well as for that of Ostia Antica, the association of
359 the ostracodal marine group with high sediment Na content unambiguously reveals a marine-
360 dominated environment (Delile et al., 2014b; Goiran et al., 2014; Sadori et al., 2016) for the
361 basal part of core PO2 (Figs. 2, 4B). This marine influence is opposed mainly to the inputs of
362 heavy metals derived from human activities (Sn, Cd and Pb). Since marine influence is not
363 associated with any trace metal pollutants, we conclude that anthropogenic pollutants were
364 carried by the freshwater of the Tiber River (Delile et al., 2017). This is supported by the
365 ostracodal freshwater group, which increases greatly in the upper part of unit B1 (Fig. 2).
366 Owing to Factor 4, we can show for the first time that the functional harbor unit B1 recorded
367 two distinct regimes (gray dotted line in Fig. 4B): a harbor basin under marine influence at a
368 depth of about 9 to 7 m *b.g.l.* (i.e., 4.6-6.6 m *b.p.s.l.* or 3.8-5.8 m *b.a.s.l.*) followed by an
369 increasingly fluvial-dominated regime at a depth of about 7 to 6 m *b.g.l.* (i.e., 3.6-4.6 m
370 *b.p.s.l.* or m 2.8-3.8 m *b.a.s.l.*).

371 4.2. *Synthesis of the main environmental factors leading to the formation of ancient harbor*
372 *basin deposits in the Mediterranean*

373 The methodological tool associating elemental geochemistry with factor analysis in
374 order to improve the reconstruction of paleo-environments has been applied to four other
375 roman harbor basins in the Mediterranean besides that of Ostia Antica used as an example
376 above: that of Ephesus in Turkey (Delile et al., 2015), that of Naples in Italy (Delile et al.,
377 2016a) and those of Trajan (Delile et al., 2014a) and Claudius (this study) at *Portus*. Table 1
378 presents the first synthesis of this method in describing the different environmental variables
379 (that is, the factors identified by factor analysis) that affected the paleo-environmental
380 dynamics of the water columns of these ancient harbor basins, as well as their respective
381 weights expressed in percentages. F1 corresponds to the most important control factor, often
382 representing around 50% of the variance, while F4 constitutes the least significant
383 environmental variable with values of less than 10% (Table 1).

384 According to Table 1, the first environmental component (42%) that affects the paleo-
385 environmental dynamics of harbor water columns corresponds to detrital fluxes, that is,
386 sedimentary discharge from an outlet. This is not surprising as all these harbor sites, except
387 that of Naples, are located on deltas that are sub-aqueous and sub-aerial coastal accumulations
388 of river-derived sediments. The process of construction of these complex coastal landforms is
389 based on the terrigenous detritus of fluvial origin, the effectiveness of which increases at river
390 mouths, where the ancient harbor basins are located. This principal environmental
391 characteristic of harbor environments is also present in the typology of ancient harbors
392 produced by Giaime et al. (2016). According to the statistical applications (Principal
393 Component Analysis and Hierarchical Ascendant Classification) on which their typology
394 rests, the major natural process that distinguishes between specific types of harbors is the rate
395 of sediment supply. This process moreover is responsible for the major constraint on

396 navigation as it reduces the draught for ships in the harbor basins because of siltation and
397 alluviation (Goiran and Morhange, 2003; Marriner and Morhange, 2007; Salomon et al.,
398 2016; Goiran et al., 2017). The predominance of detrital fluxes in the paleo-environmental
399 evolution of the water column in harbor basins is subject to the coastal geomorphology of the
400 sites where these harbors were established, as only those located at deltas are dominated by
401 this environmental parameter. Indeed, the paleo-environmental changes of the water column
402 of the ancient harbor of Naples, located in a small coastal bay, were affected more by aeration
403 (Factor 1 = redox conditions = 36%) than by terrigenous contributions (Factor 2 = 16%) (see
404 Table 1 and/or Delile et al., 2016a). In general, these detrital fluxes are composed of
405 siliciclastic and aluminosilicate minerals which cause enrichment in Al, Zr, Ti, Th, Mg, Ba,
406 K, Co, Zn, and REEs in the sediments. This category of flux generally reflects the ambient
407 hydrodynamics of harbor basins in which the recrudescence of currents is produced either by
408 the activity of canals connecting the harbor basins to the river (in freshwater-dominated
409 harbor environments), or by the acceleration of deltaic progradation (on floodplains, coastal
410 alluvial fans, paleo-channels, delta-fronts, and prodeltas) (Table 1).

411 Generally, these allochthonous particle fluxes are anti-correlated with the second source
412 of sedimentary particles in harbor deposits, the biogenic fluxes (34%, Table 1). This negative
413 correlation is due to the nature of these fluxes which are considered autochthonous as they are
414 produced *in situ*. This is why this environmental variable is so predominant in the water
415 columns of the harbor basins of Trajan and Ephesus (Table 1). The grouping of elements
416 representative of biogenic fluxes (L.O.I., Ca, and Sr) is usually found on the axis of Factor 1,
417 in a position opposite to that of the constituent elements of detrital fluxes. They are most often
418 evidence for relatively calm hydrodynamic conditions as these favor the production of
419 biomass, whether plant (organic matter) or carbonated (skeletal remains). While the
420 production of biomass depends on paleo-geographic changes that in turn encourage

421 environments with weaker water currents (swamp, marine, enclosed harbor, peat, lagoon,
422 epilimnic, brackish, or eutrophic), eutrophication and urban sewage inputs also participate
423 effectively in the production of these biogenic fluxes.

424 In order of importance, authigenic fluxes (20%) are next; these correspond to the redox
425 conditions of the environment. This environmental parameter expresses the conditions of the
426 ventilation of the harbor water column, that is, its speed of renewal, which is rapid for well-
427 oxygenated milieus (Mn, K, Ti, Zn, Cr, Co, Cd), and slow in environments deficient in
428 oxygen (S, Mo, U, Pb, Ag, Cu). In the first case, deltaic progradation (on floodplains,
429 freshwater-dominated harbors, coastal alluvial fans, paleo-channels, delta-fronts and
430 prodeltas) and breaking of waves on the shore (on delta-fronts and beaches) are the principal
431 processes of oxygenation identified. In the second case, the processes limiting the presence of
432 dissolved oxygen in the harbor waters are peat formation arising from enclosing harbor
433 basins, interflood periods, urban sewage inputs, and eutrophication (in enclosed harbors and
434 peat bogs). The range of anoxic environments is relatively extensive, as we have identified
435 marine, lagoon, marshy, peaty, and harbor environments. In terms of paleo-environmental
436 reconstructions, this factor is particularly instructive for evaluating the degree to which the
437 environments encountered are open; the more the milieu is open, the higher the proportion of
438 dissolved oxygen, and vice versa.

439 The following factor, which can sometimes be intermixed with Factor 1, expresses the
440 mineralogy and the lithology of the sediments (11%). This factor therefore characterizes
441 rather well the granulometric distribution of the stratigraphic sequences studied, in which the
442 fine sediments are dominated by alumino-silicates (Al, Ti, Mg, Rb, Ga, Li) and the coarsest
443 by siliciclastic minerals (Zr, Th, REEs, Si). Besides this characteristic useful for
444 reconstructing variations in the hydrodynamics of harbor currents, this environmental variable

445 also provides unique information on the geological sources of sediments from a given
446 watershed.

447 The fifth variable revealed by factor analysis represents the pollutants of anthropogenic
448 origin (10%) (Table 1). This component is made up of heavy metals (Pb, Cu, Sn, Ag, Cd)
449 which have been exploited by humans in various ways for millennia. However, it is necessary
450 to be cautious in using this factor because the precipitation and thus the enrichment of these
451 metallic trace elements depends on the intensity of human activity, but also on the physico-
452 chemical conditions (Delile et al., 2015) and the turbulence of the aquatic milieu (Delile et
453 al., 2017). In order to be free of these two environmental constraints that can bias the reading
454 of human influence over time, we normalize the trace elements of interest to an element
455 representative of the terrestrial crust (Al, Th, REEs, etc.) and calculate the enrichment factor
456 on the basis of representative crustal values specific to the local context in question. These are
457 easily obtainable from the pre-harbor levels of the studied sites.

458 The last factor of environmental control, which completes the total geochemical
459 variability in a given harbor deposit sequence, is the marine influence (10%) in the sediments
460 based on their enrichment in Na (Table 1). This factor is particularly useful when the absence
461 of microfauna renders reconstructions of paleo-environments difficult. Generally in the case
462 of a harbor milieu open to the sea, the increase in the proportion of seawater compared to
463 freshwater leads to the formation of a harbor milieu under marine influence (i.e., a marine-
464 dominated harbor). A large range of harbor facies depends on the degree of opening of the
465 harbor basin to the sea and its distance from the river mouth (if present). In contrast, an
466 increase in the values of this factor generally characterises pre-harbor levels of the delta front
467 type.

468 Once these environmental variables have been identified, combining their respective
469 information enables determination of paleo-environments and the processes that produced
470 them.

471 *4.3. The sources of the Tiber river-mouth sediments inferred from Pb isotopes*

472 Within the scope of previous studies focusing on lead pollution generated by Rome and
473 trapped in (recorded by) its harbor basins during the Roman and medieval periods (Delile et
474 al., 2014b, 2017), Pb isotope compositions were measured on four harbor sediment cores. In
475 the following, we proceed with an in-depth study of the natural Pb isotopic signal to
476 understand its meaning. Of the total of 177 samples analysed, 75 were considered
477 uncontaminated (Figs. 5 et 6). The Pb isotopic compositions of these samples reflect the
478 natural background of Pb in the waters of the Tiber whose characteristics are related to the
479 geology of the Tiber River watershed.

480 In order to optimize the reading of the geological signal from the Pb isotopes, we
481 converted the Pb isotope compositions into their corresponding geochemically informed
482 parameters, which are the Pb model age T_{mod} , the $^{238}\text{U}/^{204}\text{Pb}$ (μ) and the $^{232}\text{Th}/^{238}\text{U}$ (κ) ratios
483 (Fig. 5B) (Albarède et al., 2012). In short, T_{mod} (Ma = million years) is a proxy tracking the
484 tectonic age of crystalline rocks and their associated ore deposits, and thus closely maps the
485 distribution of the Alpine, Hercynian, and early Paleozoic provinces of Europe. μ (μ) and
486 κ (κ) delineate collision belts or tectonically stable areas, and thus divide Europe into
487 coherent regions (Blichert-Toft et al., 2016).

488 In Fig. 5, uncontaminated harbor sediments from Ostia Antica (core PO2) and Portus
489 (cores TR14, CN1, and PTXI-3) include one main source of natural Pb labeled “component
490 α ” (Delile et al., 2014b, 2017). This natural component α consists of two sub-components α'
491 and α'' connected by a mixing line (grey dotted line in Fig. 5). As recently demonstrated by
492 Delile et al. (2014b, 2017), these uncontaminated sediments represent a mixture of two local

493 low- $^{204}\text{Pb}/^{206}\text{Pb}$ sources, reflecting the geology of the Tiber River watershed. One source is
494 the upstream, erodible recent Apennine limestones (component α''), which are very similar to
495 Pb dissolved in modern Mediterranean seawater (Stumpf et al., 2010), and the other is the
496 recent volcanic rocks of the Alban Hills (component α') (Conticelli et al., 2002; D'Antonio et
497 al., 1996). As mentioned in section 4.1.2., the elemental composition of the volcanic rocks of
498 the Alban Hills is similar to that of the RMP-LD. This geochemical homogeneity of the
499 volcanic districts located in the vicinity of the Tiber Delta (Colli Albani, Vulsini, Cimini,
500 Vicano, Ceriti, and Sabatini) is also observed for their Pb isotopic compositions (Conticelli et
501 al., 2007; Mattei et al., 2010). In other words, the Pb isotopic fingerprint of the Alban Hills
502 (component α') reflects that of the RMP-LD as a whole. Furthermore, in the trend $\alpha'-\alpha''$
503 uncontaminated sediments split distinctly into two groups according to their kappa values
504 (Fig. 5A) or $^{208}\text{Pb}/^{206}\text{Pb}$ ratio (Fig. 5B). The sandy pre-harbor sediments show high values,
505 while the harbor sediments dominated by clays and silts are characterized by lower values. It
506 thus appears that a particle size gradient can be inferred from the Pb isotopic compositions of
507 the sediments.

508 To test this linear relationship, we calculated two indices labeled $f\alpha'$ and $f\alpha''$ that
509 measure the proportion of the natural sub-components α' and α'' in all uncontaminated
510 sediment samples (equation details in the Materials and Methods section of Delile et al.,
511 2017). In Fig. 6, where these index values are plotted against the sand, silt, and clay
512 percentages of the same samples, it is clear that the particle size distribution of the Tiber delta
513 sediments largely correlates with the proportions of the two main natural sedimentary sources
514 of the Tiber's watershed. On the one hand, sandy particles correlate positively with the
515 volcanic component ($f\alpha'$) ($r = 0.69$) and, on the other hand, fine particles correlate well with
516 the Apennine limestones ($f\alpha''$) ($r = 0.68$) (Fig. 6). Such a distinction is not unexpected, as the
517 changes in mineral sorting processes that result from varying levels of hydrodynamic activity

518 modify the mineralogy and thus the geochemistry of the transported sediments. According to
519 Garçon et al. (2014), a “heavy mineral effect” alters the Pb isotopic budget of both coarse and
520 fine fluvial sediments. Some heavy minerals concentrated in coarse sediments are extremely
521 radiogenic, allowing sandy sediments to grow in more radiogenic $^{208}\text{Pb}/^{206}\text{Pb}$ values (Garçon
522 et al., 2014) and, thereby, acquire elevated values of κ (r between $^{208}\text{Pb}/^{206}\text{Pb}$ vs $\kappa \sim 0.7$). The
523 sandy deposits of the Tiber delta's littoral facies are strongly enriched in heavy minerals
524 (Belfiore et al., 1987) that take the form of placers (Salomon, 2013) (Fig. S3) derived from
525 the erosion of the Monti Sabatini or the Colli Albani (Funicello, 1995; Ventriglia, 1971). The
526 presence of such minerals requires a source close to the deposit area (Carling and Breakspear,
527 2006), located less than 20 km away according to Thomas and Thorp (1993).

528 A primarily volcanic origin for the sandy particles of the Tiber delta, which would have
529 derived from erosion of the volcanic rocks in the Alban Hills, but also from the other volcanic
530 districts that are part of the RMP-LD surrounding the Tiber delta (Vulsini, Cimini, Vicano,
531 Ceriti, and Sabatini), is reinforced by a particularly significant correlation coefficient between
532 F1 and fa' measured in core PO2 ($r = -0.7$).

533 4.4. Chronology of the life cycle of the harbor

534 According to the present sedimentological, geochemical, and Pb isotopic results, the
535 paleo-environmental evolution of the water column of Rome's first harbor took place in five
536 stages (gray and white bands in Fig. 4B) as described further below. First, however the
537 chronostratigraphic framework of the PO2 core will be discussed.

538 4.4.1. Comments on the chronostratigraphic model of the PO2 core

539 The S-shape of the ^{14}C age-depth model obtained for the PO2 core (Fig. 2) testifies to a
540 well-known and well-documented phenomenon in harbor geoarcheology resulting from
541 multiple dredging operations. These practices, observed in many ancient Mediterranean

542 harbor basins, consist in removing part of the sedimentary thickness of the harbor deposits in
543 order to preserve a sufficiently deep draught to guarantee the circulation of ships and thus
544 maintain the operation of the port. This phenomenon is generally detected on the basis of (1)
545 sharp drops in apparent sedimentation rates and (2) chronological inversions due to removal
546 and mixing of part of the harbor deposits (Marriner and Morhange, 2006). At Ostia, such
547 dredging markers have been identified several times and have been the subject of specific
548 studies by Salomon et al. (2016) and Goiran et al. (2017). To sum up these studies, the
549 chronostratigraphic model of the ancient port basin of Ostia contains four chronological gaps
550 reflecting three potential dredging phases. These gaps are distributed as follows (red stars in
551 Fig. 2):

552 **Gap 1** is identified by a sharp drop in sedimentation rate between the last date from unit
553 A at 1050 cm depth (835-736 BC) and the first date from unit B1 at ~ 870 cm depth (358-113
554 BC). This chronological gap either testifies to the excavation of the harbor basin of Ostia or to
555 a later dredging phase that occurred around the middle of the 4th c. BC.

556 **Gap 2** is identified by a chronological inversion recorded at 815 cm depth (728-265
557 BC) reflecting the mixing of part of the harbor deposits where older deposits are mixed in
558 with younger deposits. Since this date lies within the chronological ranges of the sedimentary
559 units A and B, this older date may refer to the same dredging event described above. Another
560 explanation could be that this date, which is based on terrestrial material (charcoal), was part
561 of reworked material coming from the Tiber.

562 **Gap 3** is identified by a sharp drop in the sedimentation rate between the last date of
563 unit B1 at ~ 585 cm depth (387-204 BC) and the first date from unit B2 at 526 cm depth (163
564 BC-AD 16). This chronological gap reflects a second potential dredging phase that took place
565 at the earliest before the mid-2nd c. BC.

566 **Gap 4** is identified by a last sharp drop in the sedimentation rate between the date at 378 cm
567 depth (158 BC-AD 24) and that at ~ 550 cm depth (AD 80-231). This chronological gap
568 reflects a third and last potential dredging phase that took place at the latest during the 1st c.
569 AD.

570 4.4.2. *The pre-harbor environment*

571 The pre-harbor unit A is composed of bedded and shelly grey sands with *Posidonia*,
572 attesting to a depositional environment of deltaic progradation. According to the age-depth
573 model of the core (Fig. 2), this unit dates from the early 1st millennium (modeled dates are
574 situated between the end of the 9th c. and the early 8th c. BC) to the middle of the 4th c. BC
575 (Goiran et al., 2014, 2017; Delile et al., 2017). Figure 4 shows a relatively high-energy
576 environment revealed by the negative values of Factor 1, faithfully reporting the rather coarse
577 grain size of this unit (Fig. 2). The main geochemical aspects of this unit are the high salinity
578 (high Na, F4 in Fig. 4B) and the oxygen depletion (negative values of the Factor 3 in Fig. 4B),
579 consistent with the marine-to-brackish lagoonal ostracod groups observed by Goiran et al.
580 (2014). Also reported in this work are freshwater inflows (Fig. 2) that explain the turbulent
581 conditions attested to by the siliciclastic flux (Factor F1). While the main mineralogical and
582 geochemical characteristics of these pre-harbor deposits are like those recorded at *Portus*
583 (Delile et al., 2014a), the freshwater influence on the site of the ancient harbor basin of Ostia
584 Antica strengthens the hypothesis of the presence of an estuarine river mouth, whose fluvial
585 sand inputs were remobilized by the marine swell. Due to the continuity and regularity of the
586 seawater influence during almost five centuries, as well as the dramatic oxygen depletion (F3
587 negative values in Fig. 4) and the freshwater influence, it is difficult to posit this pre-harbor
588 deposit as a credible "tsunami event I" as has been postulated between 800 BC and 400 BC by
589 Hadler et al. (2015a). In the somewhat more realistic and less catastrophic perspective taken
590 here, the assemblage of observed and measured faunal, sedimentological, and geochemical

591 indicators rather converge towards a pre-harbor deposit typical of a classic deltaic front
592 sequence, which is well recorded both at the scale of the site itself and of the Tiber delta as a
593 whole (Mazzini et al., 2011; Goiran et al., 2012; 2014; Salomon, 2013; Salomon et al., 2012,
594 2018; Delile et al., 2014a; Sadori et al., 2016)

595 4.4.3. The lower harbor sequence

596 From the middle of the 4th c. to the beginning of the 2nd c. BC, the environmental
597 conditions change markedly towards a quiet milieu between -9 and -5.7 m *b.g.l.* (i.e., between
598 6.6 and 3.3 m *b.p.s.l.* or between 5.8 and 2.5 m *b.a.s.l.*) (Fig. 4), as suggested by the compact
599 dark grey silt deposits (Goiran et al., 2012, 2014, 2017; Sadori et al., 2016). Classically, this
600 unit B1 resulted from the creation of the harbor basin of Ostia Antica, a typical well-protected
601 harbor, still connected to fluvial-marine environments. The significant reduction of the
602 hydrodynamic conditions of the currents leading to the deposit of unit B1 is clearly indicated
603 by Factor 1, which shows a drastic decrease in siliciclastic fluxes (Fig. 4B). Inversely, the
604 same process led to an increase in the proportion of fine particles in the deposit as seen in the
605 positive values of Factor 2 (aluminosilicate flux in Fig. 4B). The third geochemical
606 characteristic of unit B1 is the distinction of two regimes in the paleo-environmental
607 dynamics of the harbor water column. Based on the seawater influence (Factor 4 in Fig. 4B),
608 unit B1 can be divided into two subunits:

- 609 - Subunit B1a (from -9 to -7 m *b.g.l.*; i.e., between 6.6 and 4.6 m *b.p.s.l.* or between 5.8
610 and 3.8 m *b.a.s.l.*) reflects a marine-dominated regime since the F4 values (seawater
611 influence) always remain positive, although being lower than those of unit A due to the
612 connection of the harbor basin to the Tiber river;
- 613 - Subunit B1b (from -7 to -5.7 m *b.g.l.*; i.e., between 4.6 and 3.3 m *b.p.s.l.* or between 3.8
614 and 2.5 m *b.a.s.l.*) reflects a freshwater-dominated regime since the F4 values (seawater

615 influence) are negative. This is corroborated by the ostracodal freshwater group in this
616 subunit (Fig. 2).

617 According to the ^{14}C age-depth model of the PO₂ core (Fig. 2A), the passage from
618 subunit B1a to B1b took place around the middle of the 3rd c. BC. This change in the paleo-
619 environmental dynamics of the harbor water column at this particular time is not surprising,
620 and even rather consistent with the known progradation phases of the Tiber river delta.
621 [Giraudi et al. \(2009\)](#), [Bicket et al. \(2009\)](#), [Giraudi \(2011\)](#), and [Salomon et al. \(2018\)](#) recorded
622 intense Tiber delta progradation between the 4th c. BC and the 5th c. AD which began after a
623 sudden paleo-geographic event. Indeed, a major step of the Tiber delta construction occurred
624 between the 8th and the 3rd c. BC due to the sharp southward migration of the river mouth, in
625 the direction of the harbor basin of Ostia ([Bellotti et al., 2011](#); [Milli et al., 2013](#); [Salomon et](#)
626 [al., 2018](#)). Over this time period, the rate of the progradation of the river mouth is estimated to
627 be around 5-6 m/year ([Milli et al., 2013](#)). According to [Milli et al. \(2013\)](#) a strong Tiber flood
628 event that occurred during the climate cooling phase of the Iron Age (Bond cycle B2, [Bond et](#)
629 [al., 1997, 2001](#)) probably led to the shift of the Tiber mouth location. In this new paleo-
630 geographical context of the Tiber delta, the increasing fluvial inputs into the harbor basin of
631 Ostia Antica between the 4th and the 2nd centuries BC (the chronological range of unit B1)
632 caused the paleo-environmental dynamic change of the harbor water column towards a
633 freshwater-dominated regime in about the early 3rd c. BC.

634 This gradual and continuous increase of the freshwater influence in the harbor muds is
635 not consistent with the surge of a tsunami deposit in the “lagoonal harbour” which is referred
636 to as “event II” by [Hadler et al. \(2015a\)](#). Such a “high-energy event layer” should present a
637 sudden break and a sustainable return of seawater influence in this deposit, and not the
638 reverse.

639 *4.4.4. The upper harbor sequence*

640 Between the 2nd c. BC and the 3rd c. AD, a new harbor sequence called unit B2 became
641 established at a depth of between 5.7 and 3.2 m *b.g.l.* (i.e., between 3.3 and 0.8 m *b.p.s.l.* or
642 between 2.5 and 0 m *b.a.s.l.*; Figs. 2 and 4). This unit presents sedimentological and
643 geochemical characteristics that are different from the lower harbor sequence as the deposit of
644 coarse sands (Fig. 2) led to a retreat of aluminosilicate minerals (negative values of Factor 2)
645 in favor of siliciclastic particles (negative values of Factor 1). In unit B2, three major pulses
646 of siliciclastic flux reflect periods of strong hydrodynamic currents in the harbor basin; they
647 are evidence of high-energy floods of the Tiber (Goiran et al., 2014, 2017, Salomon et al.,
648 2016). These phases were followed by periods of relatively calm currents in which anoxic
649 conditions were established (negative values of Factor 3 in Fig. 4B). Finally, unit B2 reflects
650 infilling of the harbor basin, which appears to have been subject to the hydrological rhythms
651 of the Tiber. During periods of major inundations, the currents renewed the oxygenation of
652 the water masses, while in periods of low water level, the less energetic currents led to the
653 development of anaerobic conditions.

654 According to Hadler et al. (2015a) and Wunderlich et al. (2018) unit B2 would
655 incorporate yet a third tsunami event dated to between AD 1 and 50. As speculated above for
656 their second tsunami event, our geochemical analyses should show a sudden break illustrating
657 a sustainable return of seawater influence in this deposit. However, exactly the opposite is
658 recorded, since the influence of seawater is becoming increasingly absent in unit B2 (Fig. 4B).
659 Moreover, the occurrence of a tsunami would have been observed at different places in the
660 delta, such as the Ostia lagoon (Bellotti et al., 2011; Vittori et al., 2015) and the Maccarese
661 lagoon (Bellotti et al., 1994, 2007; Giraudi, 2002, 2004, 2011; Di Rita et al., 2010; Jouannic et
662 al., 2013), since the sediment-transport distances for tsunamis vary from several hundred
663 meters to a few kilometres from the shore (a regional signature) (Minoura et al., 1997;
664 Gelfenbaum and Jaffe, 2003; Morton et al., 2007). However, none of the studies carried out

665 on the sedimentary deposits of the Tiber delta have mentioned any such event. This is not
666 surprising as the three high-energy events of tsunamigenic origin were recorded in a single
667 core (core OST 3), among eight cores taken by [Hadler et al. \(2015a\)](#). Thus the tsunamigenic
668 fingerprint appears to be local rather than regional. However, as pointed out by [Sadori et al.](#)
669 [\(2016\)](#), if such a catastrophic event had taken place during the 1st c. AD, no doubt it would
670 have been recorded by Roman historians, who paid close attention to natural phenomena; this
671 is, however, not the case. Rather than assuming a scenario including a third tsunami event as
672 speculated by [Hadler et al., \(2015a\)](#), we present below in three steps using an integrated
673 approach (i.e. combined hydro-climatic, historical, and sedimentological and geochemical
674 data) an alternative scenario that takes into account the environmental context of this period.

675 (i) During the time period covered by unit B2 (2nd c. BC to 3rd c. AD), a major hydro-
676 sedimentary event, which is characterized by an increase in the frequency of floods, occurred
677 during the period of the 1st c. BC to the 2nd c. AD approximately at several places in the
678 western Mediterranean region ([Berger and Bravard, 2012](#)). Based on the flood events of the
679 Tiber River in Rome recorded [by Bersani and Bencivenga \(2001\)](#), corroborating the work of
680 [Le Gall \(1953\)](#), such an increase in the flood frequency of the Tiber took place between
681 approximately the 2nd c. BC and the 2nd c. AD.

682 (ii) During this major hydro-sedimentary crisis, several coastal sites experienced an
683 acceleration of deltaic progradation on the Mediterranean coast due to the erosion and
684 transport of substantial amounts of sedimentary loads. This was the case for the Rhône delta
685 between the 1st c. BC and the end of the 2nd c. AD ([Arnaud-Fassetta, 2000, 2002](#)), the Argens
686 delta in eastern Provence ([Excoffon et al., 2010, 2016](#)) and the Küçük Menderes delta in
687 Turkey from the 2nd c. BC to the 1st c. AD at least ([Delile et al., 2015](#)). In the case of the Tiber
688 delta, the work of [Giraudi et al. \(2009\)](#) distinctly shows an acceleration phase of progradation
689 that occurred between the 3rd c. BC and the 3rd c. AD.

690 (iii) This rapid progression of the progradation of the deltas stimulated by a sporadic
691 deterioration of hydro-climatic conditions brought on the accelerated siltation of numerous
692 harbor basins in the Mediterranean such as that of the great Roman harbor of Narbonne in the
693 1st c. (Rescanières, 2002), and those of Frejus (Excoffon et al., 2010), Ephesus (Delile, 2014;
694 Delile et al., 2015), Naples (Delile et al., 2016a), Pisa (Benvenuti et al., 2006; Mariotti Lippi
695 et al., 2007), as well as that of Trajan at *Portus* (Delile et al., 2014a). For all these reasons, we
696 consider that at this period the acceleration of the deltaic progradation of the Tiber, partly
697 controlled by climate, but partly also by humans, moved the river mouth of the Tiber even
698 further downriver, and thus even further from the harbor basin (Fig. 2), on the one hand, and
699 mechanically caused an aggradation of the alluvial floor, on the other hand. This process thus
700 favored the evacuation of the strongest floodwaters accompanied by their sands in the harbor
701 basin of Ostia Antica. This scenario also emphasises the importance of consulting
702 contemporary sources such as the Roman author Strabo (64-58 BC / AD 21-25), who wrote:
703 “Ostia had the inability to maintain or consider a convenient sheltered harbor due to the
704 amount of Tiber sediments transported down to the seashore” (Geography, 5, 3, 5, translated
705 by H. L. Jones).

706 4.4.5. The abandonment of the harbor basin

707 The yellow bedded silts of unit C (Figs. 2 and 4) from the 3rd c. AD onward represent
708 floodplain deposits which definitively isolated the harbor basin from the sea. Unit C presents
709 the same geochemical characteristics as the floodplain deposits recorded in the uppermost unit
710 (unit C) of the Trajan basin at *Portus* (Delile et al., 2014a). In both cases, the depositional
711 environment was freshwater (negative values of F4), oxygenated conditions prevailed
712 (constant positive values of F3), and the detrital particles dominated by the aluminosilicate
713 minerals demonstrate the fineness of these sediments (high positive values of F2 and F1) (Fig.

714 4). These characteristics indicate that the abandoned harbor had finally been overrun by the
715 floodplain around five centuries before the harbor of Trajan suffered the same fate.

716 This abandonment phase of the harbor basin appears to have been mistaken with the
717 establishment of a fluvial harbor (Hadler et al., 2015) called the “younger fluvial harbour
718 phase” in Wunderlich et al. (2018). Indeed, these authors argue for a shift from a harbor of
719 lagoon type towards a fluvial harbor phase (or “harbour generation II”) located between - 0.5
720 and - 1 m *b.p.s.l.* (see Fig. 7 in Hadler et al., 2015), that is to say between - 0.2 m *b.a.s.l* and +
721 0.3 m *above a.s.l.* These values, inferred from the ancient biological sea level dated to AD
722 230-450 by Goiran et al. (2009), show unambiguously that the water column disappears at
723 these depths because the environment became terrestrial. Consequently, the establishment of a
724 river harbor at these depths is strictly impossible. For this reason, we strongly suggest using
725 ancient sea levels to characterize harbor levels and phases rather than the simple measurement
726 of present sea level.

727

728 5. Conclusions

729 The 3000-year-old geochemical record preserved in the sediment core from the ancient
730 harbor basin of Ostia Antica located in the Tiber delta has provided new insights into some of
731 the aquatic paleo-environmental conditions, insights that had not been gleaned by the
732 "classical" proxies used in the geoarcheological studies carried out in this harbor basin thus
733 far. Associated with factor analysis, elemental geochemistry applied at high resolution to
734 some forty chemical elements has enabled discrimination of the main environmental factors
735 leading to the formation of the harbor basin deposits, classified in order of importance as (i)
736 strong hydrodynamics (i.e., high-velocity currents) (the siliciclastic terrigenous signal ~ 47%),
737 (ii) quiet hydrodynamics (the aluminosilicate terrigenous signal ~ 13%), (iii) water column

738 ventilation (the authigenic fluxes ~ 10%), and (iv) seawater vs. freshwater influence (~ 7%).
739 A first comparison of these environmental variables affecting the harbor water column at
740 Ostia Antica with those identified using the same method in the ancient harbors of Trajan and
741 Claudius (*Portus*), as well as those of Ephesus and Naples, has shown that the formation of
742 the harbor basin deposits took place by means of similar geochemical fluxes, whose
743 respective weights were relatively constant from one harbor basin to the other. This
744 observation is not surprising as all these harbor sites, except for that of Naples (situated in a
745 small coastal bay north of the gulf of Naples) are located on deltas. Thus the paleo-
746 environmental evolution of the water column of the harbor basins was conditioned by the
747 coastal geomorphology of the sites where the harbors were established.

748 By focusing on sediments uncontaminated by anthropogenic lead, the study of Pb
749 isotopes has shown that it is possible to both identify the geological sources of sedimentary
750 particles of the Tiber delta and distinguish the fine from the coarser particles. The particle size
751 distribution of the Tiber delta sediments largely correlates with the proportion of the two main
752 natural sedimentary sources of the Tiber's watershed: The sandy particles correlate positively
753 with the volcanic rocks, while the fine particles are subject to rough correlations with the
754 Apennine limestones.

755 The present study also sheds new light on the chronology of the life cycle of the harbor
756 at Ostia Antica. First, the Na enrichment of pre-harbor level sediments, indicating, as at
757 *Portus*, an environment dominated by marine influence, remained when the harbor basin was
758 dug out around the middle of the 4th century BC, but to a lesser degree. Then, starting around
759 the 3rd c. BC, the initial marine-dominated regime of the harbor basin shifted towards a
760 freshwater-dominated milieu due in part to intense Tiber delta progradation caused by a short
761 phase of climate deterioration, and due in part to human action. This influence of Tiber waters
762 in the water column of the harbor basin reached a threshold of no return between the 2nd c. BC

763 and the 3rd c. AD because the harbor basin was affected by the high-energy floods of the Tiber
764 river. During this period, the geochemical signal of the coarse harbor deposits carefully
765 recorded the hydrological rhythms of the Tiber, marked by the alternation of major inundation
766 phases and pronounced low-water phases. Again during this period a phase of increased
767 progradation of the Tiber delta occurred, resulting from an increase in the flood frequency of
768 the Tiber between approximately the 2nd c. BC and the 2nd c. AD. While [Hadler et al. \(2015a\)](#)
769 and [Wunderlich et al. \(2018\)](#) have advocated for a high-energy event to interpret the coarsest
770 B2 unit, our geochemical data are more consistent with the scenario proposed by [Goiran et al.](#)
771 [\(2014\)](#), [Sadori et al. \(2016\)](#) and [Salomon et al. \(2016\)](#) who call on an upsurge in the frequency
772 of floods, which carried the sands into the harbor basin of Ostia Antica that led to its
773 abandonment. This final phase of abandonment cannot be replaced by a river port
774 environment, as argued by [Hadler et al. \(2015a\)](#) and [Wunderlich et al. \(2018\)](#), since these
775 deposits are located in part above ancient sea level (- 0.2 to + 0.3 m *b.a.s.l.*); the harbor water
776 column no longer existed at that time.

777 Finally, this study has also provided the opportunity to test the hypothesis of [Hadler et](#)
778 [al. \(2015a\)](#) that argues for three high-energy events ascribed to a tsunamigenic origin, which
779 would have thrice greatly affected the harbor basin of Ostia Antica. There are many
780 arguments against this hypothesis, whether historical (absence of historical accounts of such
781 events), geomorphological (inappropriate morphology of the coastline south of the Tiber
782 River mouth), stratigraphic (no traces of tsunami deposits in the area) ([Sadori et al., 2016](#)), or
783 anthropic (highest Pb abundances (anthropic contamination) recorded in the deposits of the
784 most recent so-called tsunami) ([Delile et al., 2017](#)). While the earliest tsunami appears to have
785 been mistaken for a classic deltaic front sequence of the Tiber, the other two high-energy
786 layers should record a sudden change to a sustainable return of seawater influence in these

787 deposits. However, our multi-elemental geochemical signal records the exact opposite, a
788 strong freshwater component that increases towards the top of the stratigraphy.

789

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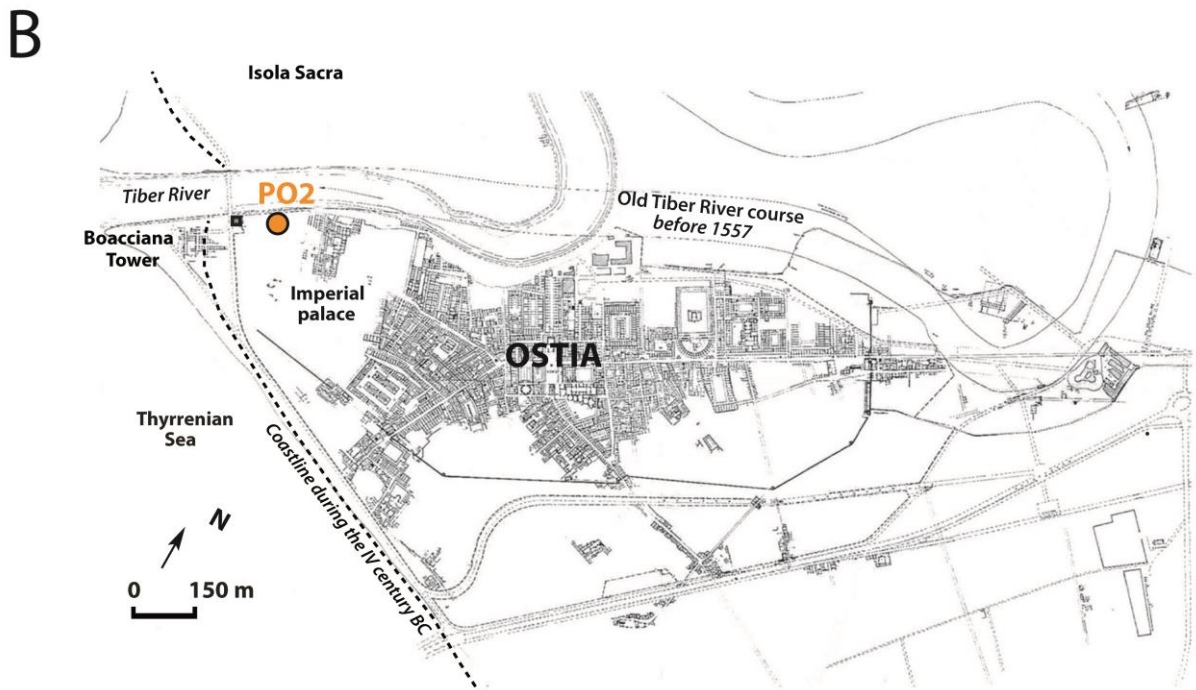
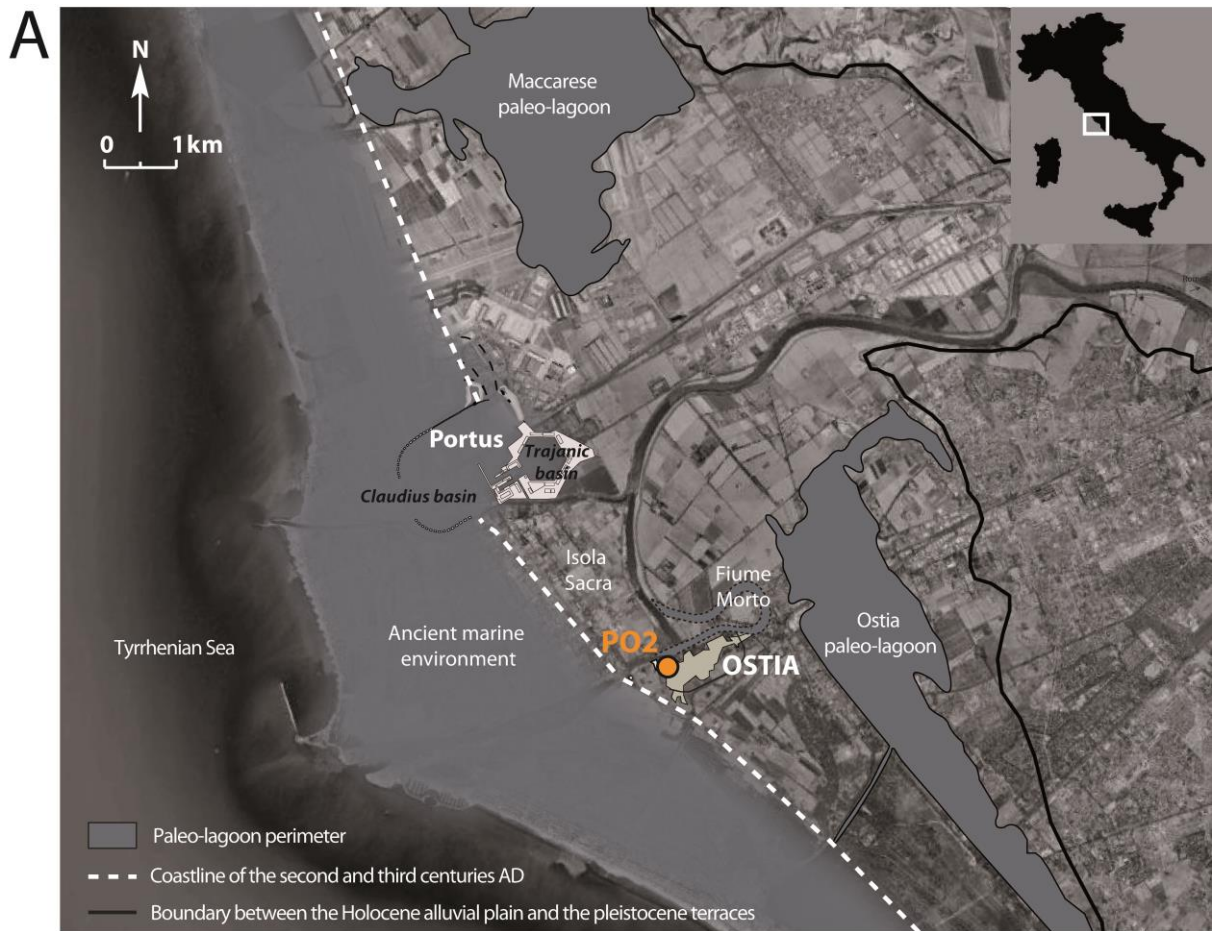
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1127 **Figure captions**

1128 **Figure 1.** (A) Location of the harbor basins of *Portus* and Ostia in the Tiber delta and position
1129 of cores PO2 and PTXI-3 (orange circles) analyzed in this work, and core TR14 analyzed by
1130 Delile et al. (2014a) (beige circle). (B) Map showing the archeological area of ancient Ostia
1131 with the location of core PO2, as well as the old (before AD 1557) and current Tiber river
1132 courses (modified from [Gorian et al., 2014](#)). The blue area is the location of the Ostia Antica
1133 harbor basin.

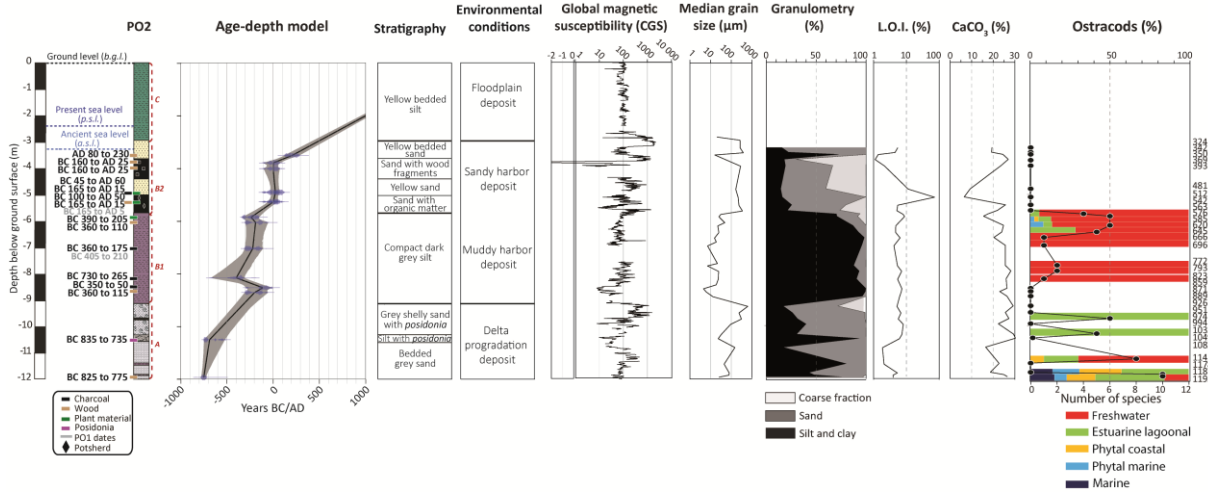


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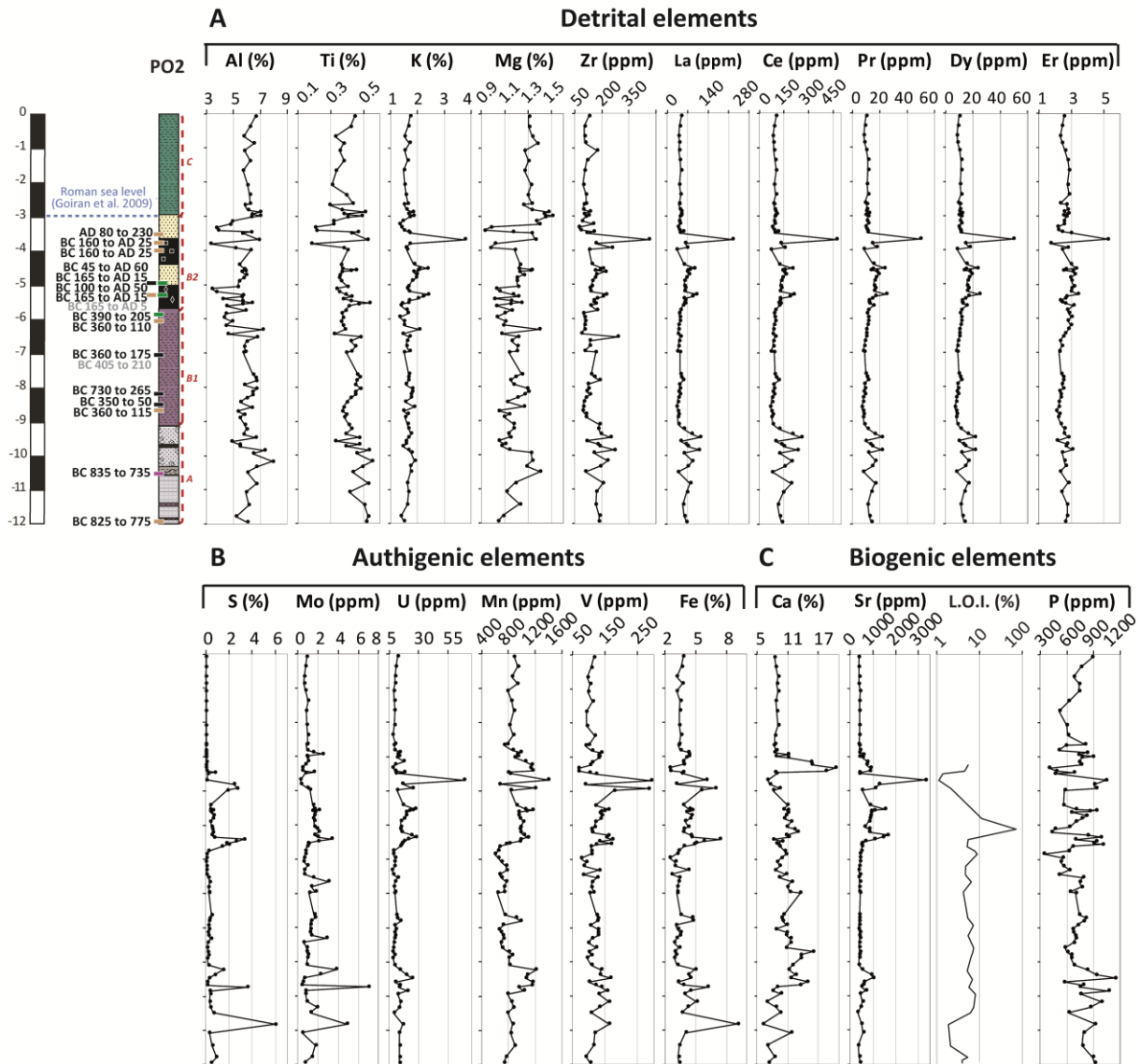
1135 **Figure 2.** Stratigraphic log of core PO2 showing the age-depth model, stratigraphic and

1136 environmental descriptions, magnetic susceptibility values, the 50 percentile (D 50) grain

1137 size, the granulometric diagram, Loss on Ignition (L.O.I.), CaCO₃ content, and ostracod
 1138 assemblages divided into five ecological groups: marine, phytal marine, coastal phytal (i.e.
 1139 phytal species requiring algae or higher plants for their life cycle), brackish lagoonal, and
 1140 freshwater (modified from Gorian et al., 2014).

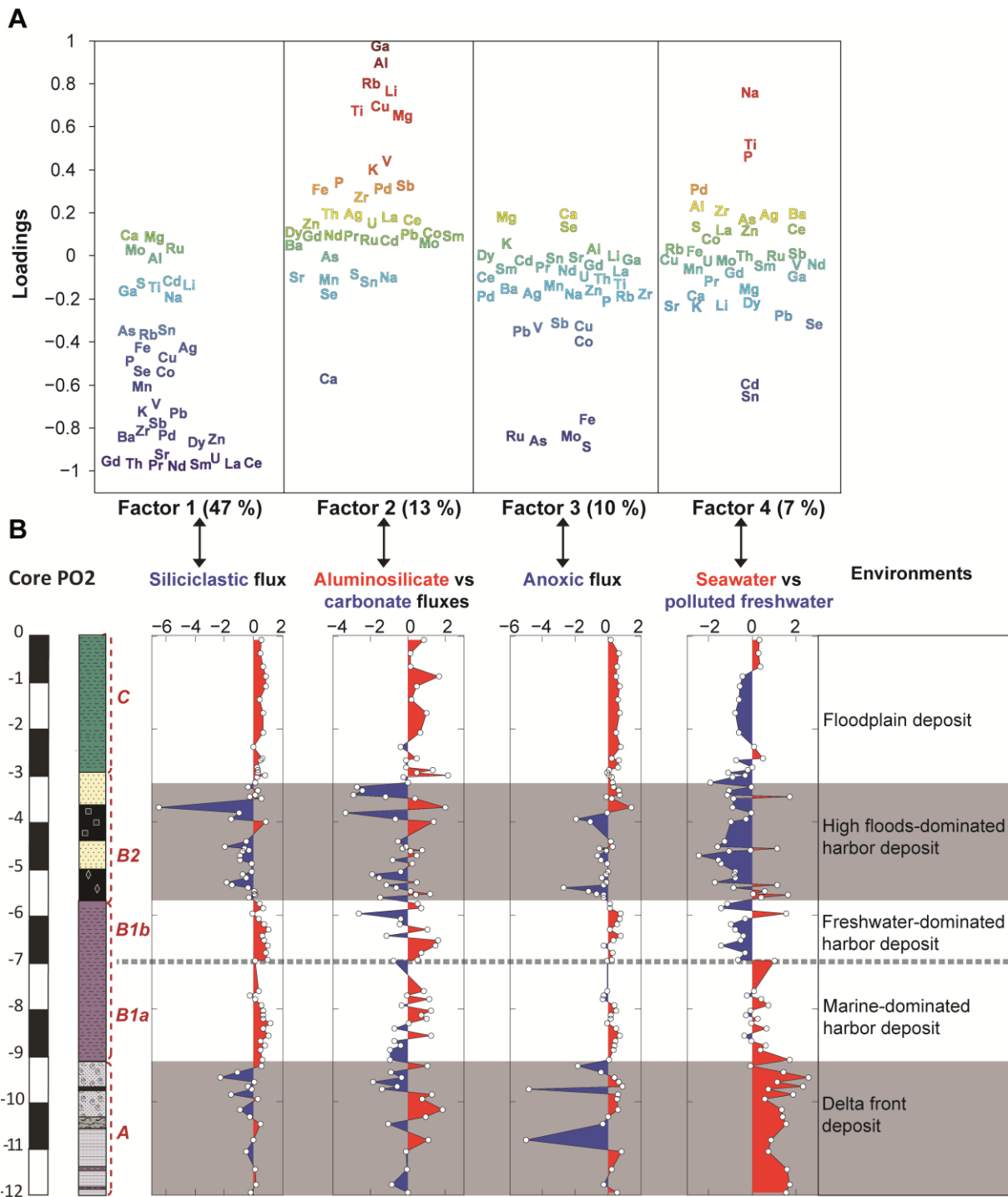


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 1142 **Figure 3.** Geochemical logs of elemental concentrations of the PO2 core classified according
 1143 to their supposed belonging to the main geochemical fluxes: (A) Detrital elements, (B)
 1144 Authigenic elements, (C) Biogenic elements. Section 4.1.1. discusses how this approach of
 1145 classifying elements according to their presumed affiliation with the main geochemical inputs
 1146 is arbitrary.



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1148 **Figure 4.** (A) Factor analysis of elemental concentrations in core PO2. The number of factors
 1149 is limited to four: (1) Silicate detrital fraction (47%); (2) aluminosilicate detrital fraction
 1150 versus carbonate fraction (13%); (3) anoxic conditions (10%); (4) seawater influence versus
 1151 polluted freshwater (7%). (B) Distribution of the different factors with depth in the column.
 1152 The gray and white shadings and the gray dotted line highlight the main geochemical units of
 1153 the core discussed in section 4.4.



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Figure 5. (A) Lead isotope ratios ($^{204}\text{Pb}/^{206}\text{Pb}$ vs. $^{208}\text{Pb}/^{206}\text{Pb}$) and (B) geochemical

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parameters (T_{mod} vs. κ) for leachates from the uncontaminated sediments of the *Trajanic* (in

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red), *Claudian* (in yellow), and *Ostia* (in green) harbor deposits, and of the *Canale Romano* (in

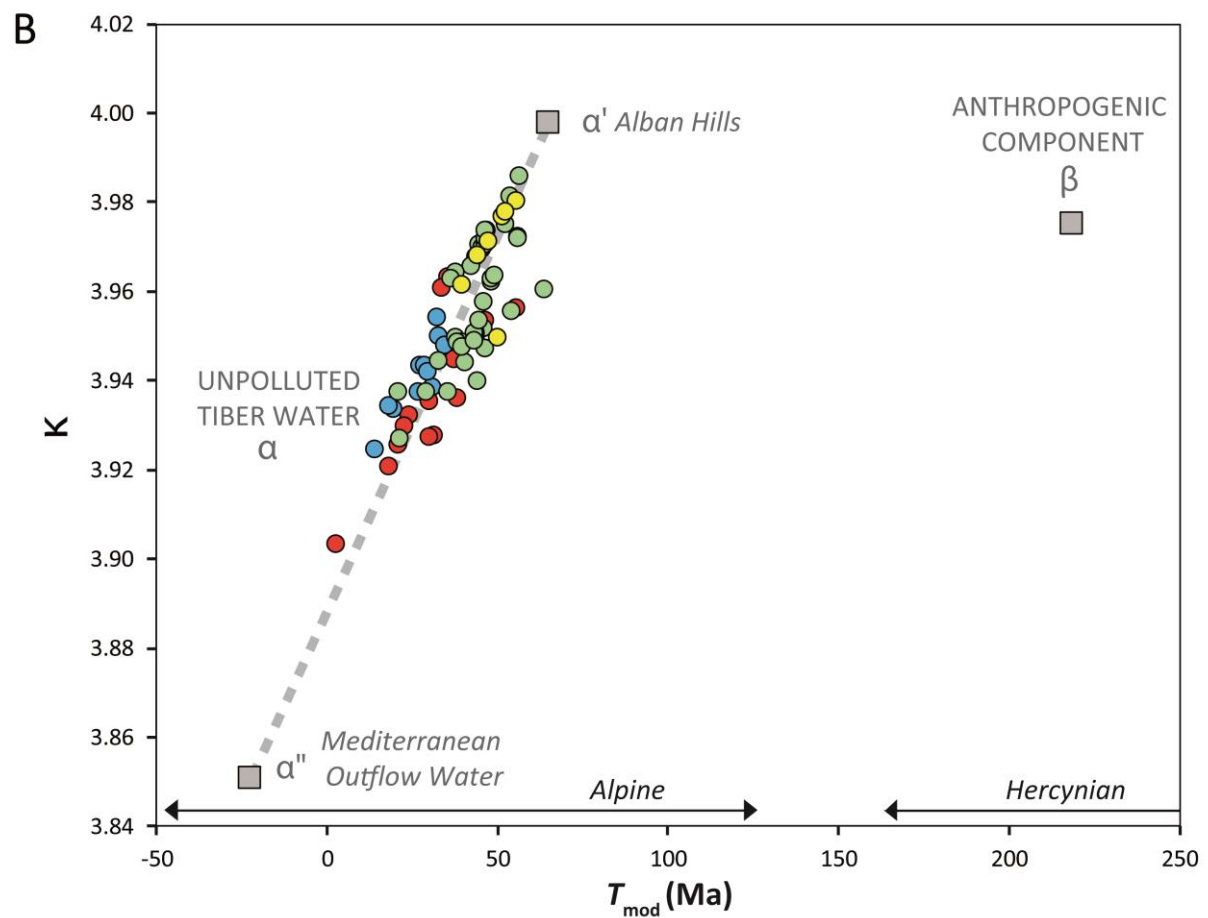
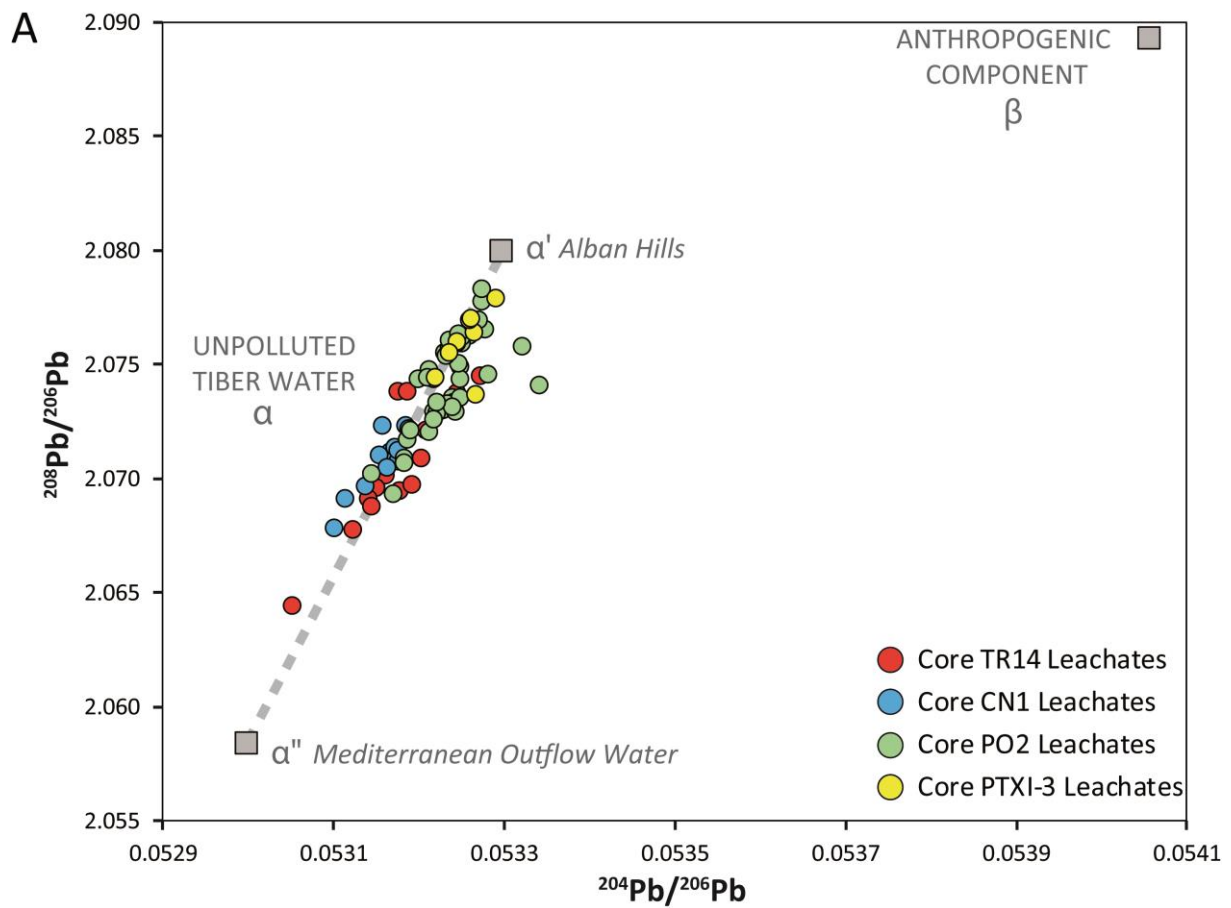
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blue). Together, these samples define a geological mixing line between the α' component

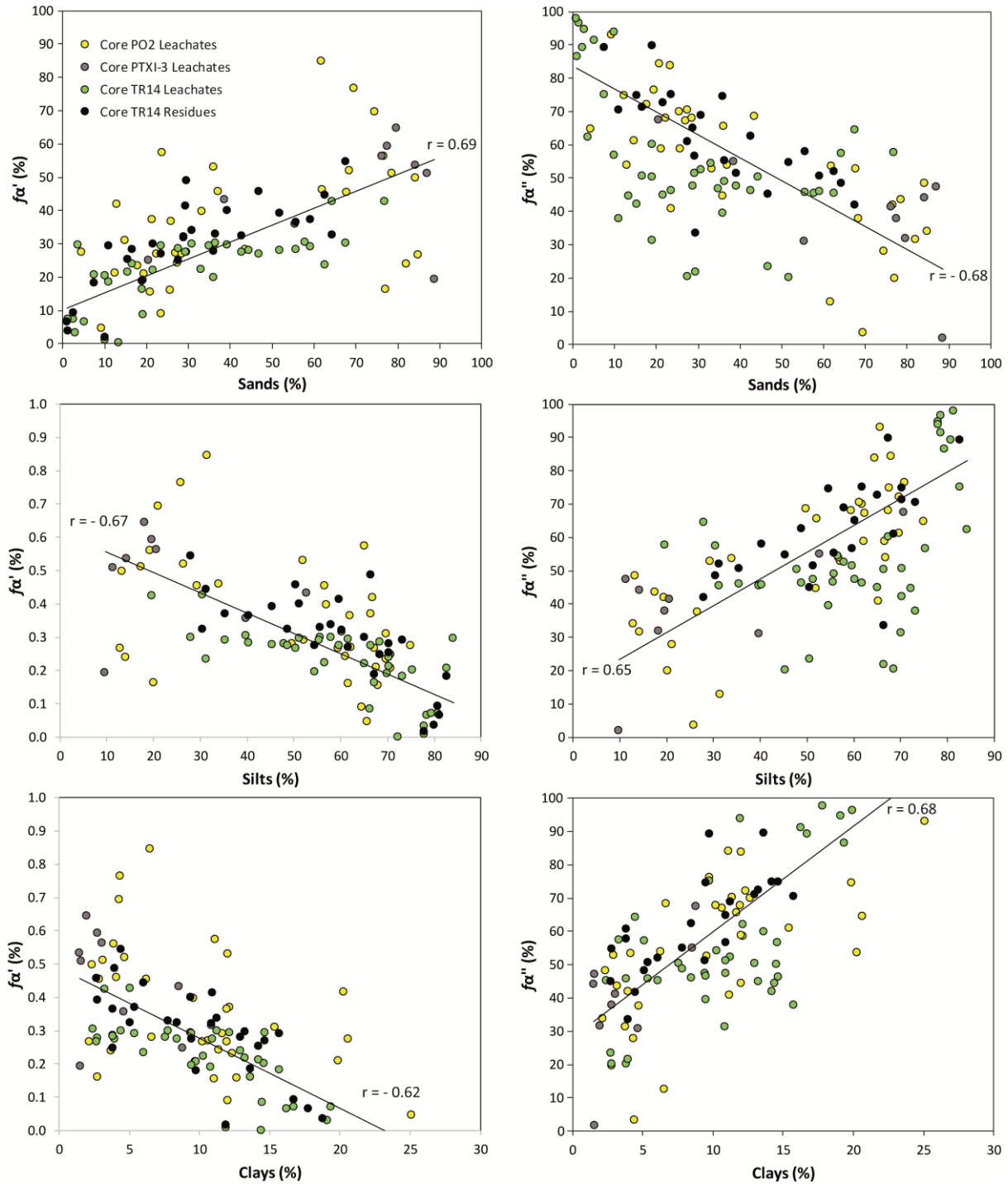
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composed of volcanic rocks from the Alban Hills (Conticelli et al., 2002; D'Antonio et al.,

1160 1996) and the α' component related to the erodible recent Apennine limestones, which are
1161 very similar to Pb dissolved in modern Mediterranean seawater (Stumpf et al., 2010)
1162 (modified from Delile et al., 2017).



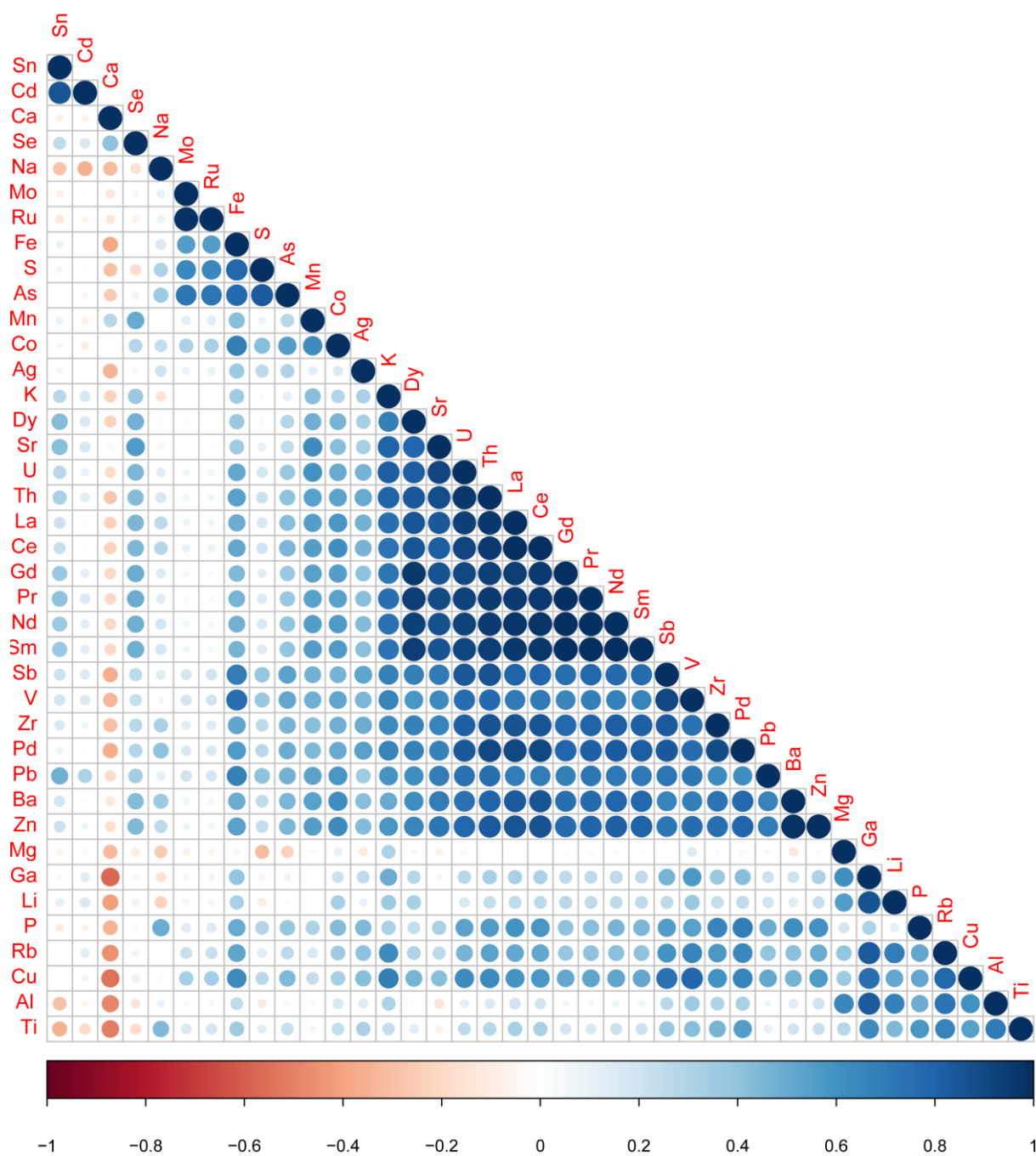
1164 **Figure 6.** Plot of $f\alpha'$ and $f\alpha''$ values (proportion of the natural sub-components α' and α'' in all
 1165 uncontaminated sediment samples) against sand, silt, and clay percentages for leachates (in
 1166 green) and residues (in black) from core TR 14 and leachates from cores PTXI-3 (in grey) and
 1167 PO2 (in yellow).



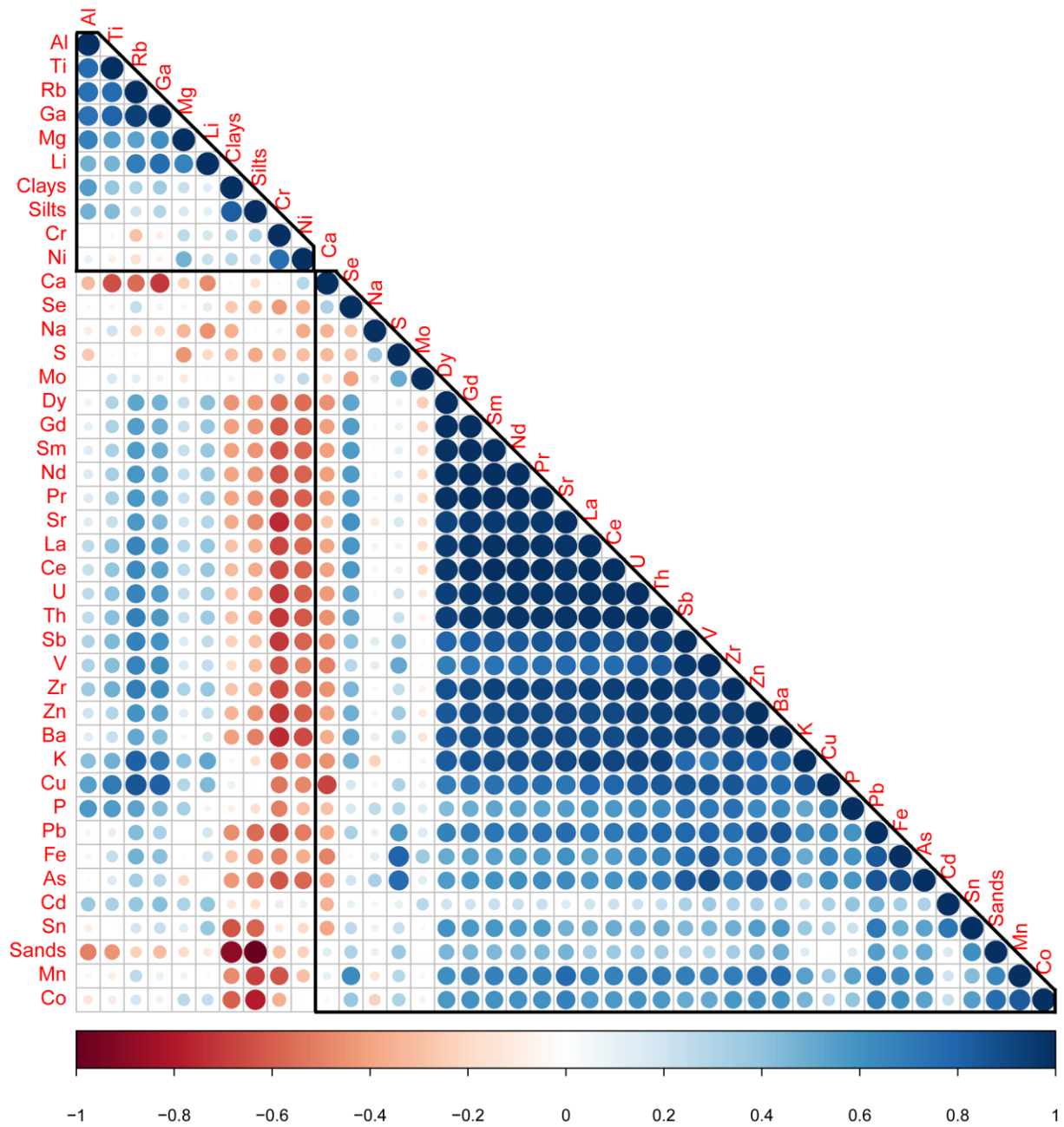
1169 **Table 1.** Environmental factors involved in the formation of sedimentary deposits in the
1170 ancient harbor basins of Ostia (Factor analysis in Fig. 4), Claudius (Factor analysis in Fig. S4
1171 and data in Table S1), Trajan, Ephesus, and Naples. These environmental variables, organized
1172 in order of relative importance, have been identified by factor analysis of element
1173 concentrations. For each of these variables, the chemical elements concerned, the associated
1174 processes, and the types of environments involved are given.

ENVIRONMENTAL FACTORS SORTED ACCORDING TO THEIR SIGNIFICANCE (F1 TO F4 IN %) AND INFERRED FROM FACTOR ANALYSIS									
Harbor site	Reference	Coastal geomorphology	Detrital flux (42%)	Biogenic flux (34%)	Oxic condition (20%)	Anoxic condition (20%)	Mineralogy/lithology (11%)	Anthropogenic flux (10%)	Seawater flux (8%)
Ostia	This study	Delta	F1 (47 %)	N/A	F3 (10 %)	F3 (10 %)	F2 (13 %)	F4 (10 %)	F4 (7 %)
Portus (Trajanic basin)	Delile et al. (2014a)	Delta	F1 (55 %)	F1 (55 %)	F2 (17 %)	F2 (17 %)	N/A	F4 (5 %)	F3 (9 %)
Portus (Claudius basin)	This study	Delta	F1 (44 %)	F2 (16 %)	N/A	N/A	F3 (12 %)	N/A	F4 (6 %)
Ephesus	Delile et al. (2015)	Delta	F1 (49 %)	F1 (49 %)	F2 (19 %)	F2 (19 %)	F3 (8 %)	N/A	N/A
Naples	Delile et al. (2016a)	Coastal bay	F2 (16 %)	F2 (16 %)	F1 (36 %)	F1 (36 %)	F3 (10 %)	F2 (16 %)	F3 (10 %)
SYNTHESIS	Significant elements	Al, Zr, Ti, Th, Mg, Ba, K, Co, Zn, REEs	LOI, Ca, Sr	Mn, K, Ti, Zn Cr, Co, Cd	S, Mo, U, Pb, Ag, Cu	Al, Rb, La, Se, Ce, Ga, Li, Be, Zr, K, Ti, Mg	Pb, Ag, Sn, Cu, Cd, Na		
	Process	Fluvial hydrodynamics: - Canal networks - Coastal and delta progradation	Biomass production: - Organic matter - Skeletal remains - Eutrophication - Urban sewage inputs	- Delta progradation - Swell	- Enclosing harbor basin - Weak water-column ventilation - Interflood period - Urban sewage inputs and eutrophication	- Erosion of the local and regional geological basement	- Heavy metal inputs - Using of lead pipe networks - Metallurgic activities	- Seawater influence - Infiltration of the littoral salt wedge	
	Paleo-environment	- Floodplain - Freshwater-dominated harbor - Coastal alluvial fan - Paleo-channel - Delta front - Prodelta environment	- Swamp - Marine - Enclosed harbor - Peat - Lagoonal - Brackish and eutrophic environments	- Floodplain - Freshwater-dominated harbor - Coastal alluvial fan - Paleo-channel - Delta front - Prodelta environments - Beach	- Marine - Enclosed harbor - Peat - Lagoonal - Swamp	In all environments	In all environments	- Marine dominated harbor - Delta front	

1176 **Figure S1.** Color-coded correlation matrix (correlogram) for all measured chemical elements
1177 in core PO2.



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1180 **Figure S2.** Color-coded correlation matrix (correlogram) for all measured chemical elements
1181 and granulometric fractions in the harbor subunits B1 and B2 of core PO2. Two groups have
1182 been identified by hierarchical clustering.

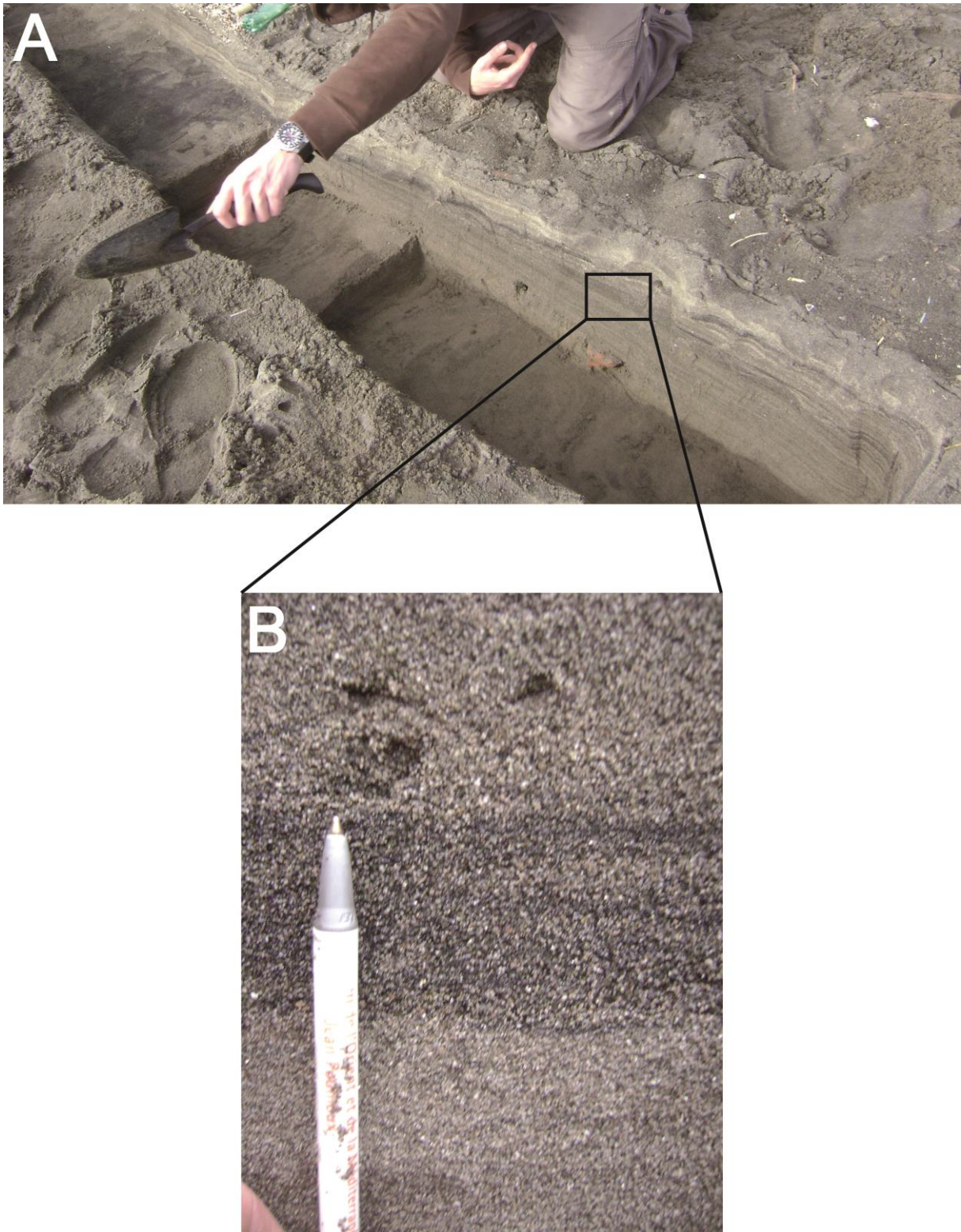


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1184 **Figure S3. (A)** Photo showing subhorizontal bedding formed by placers on the current

1185 coastline of the Tiber delta (modified from [Salomon, 2013](#)). **(B)** Close-up of a

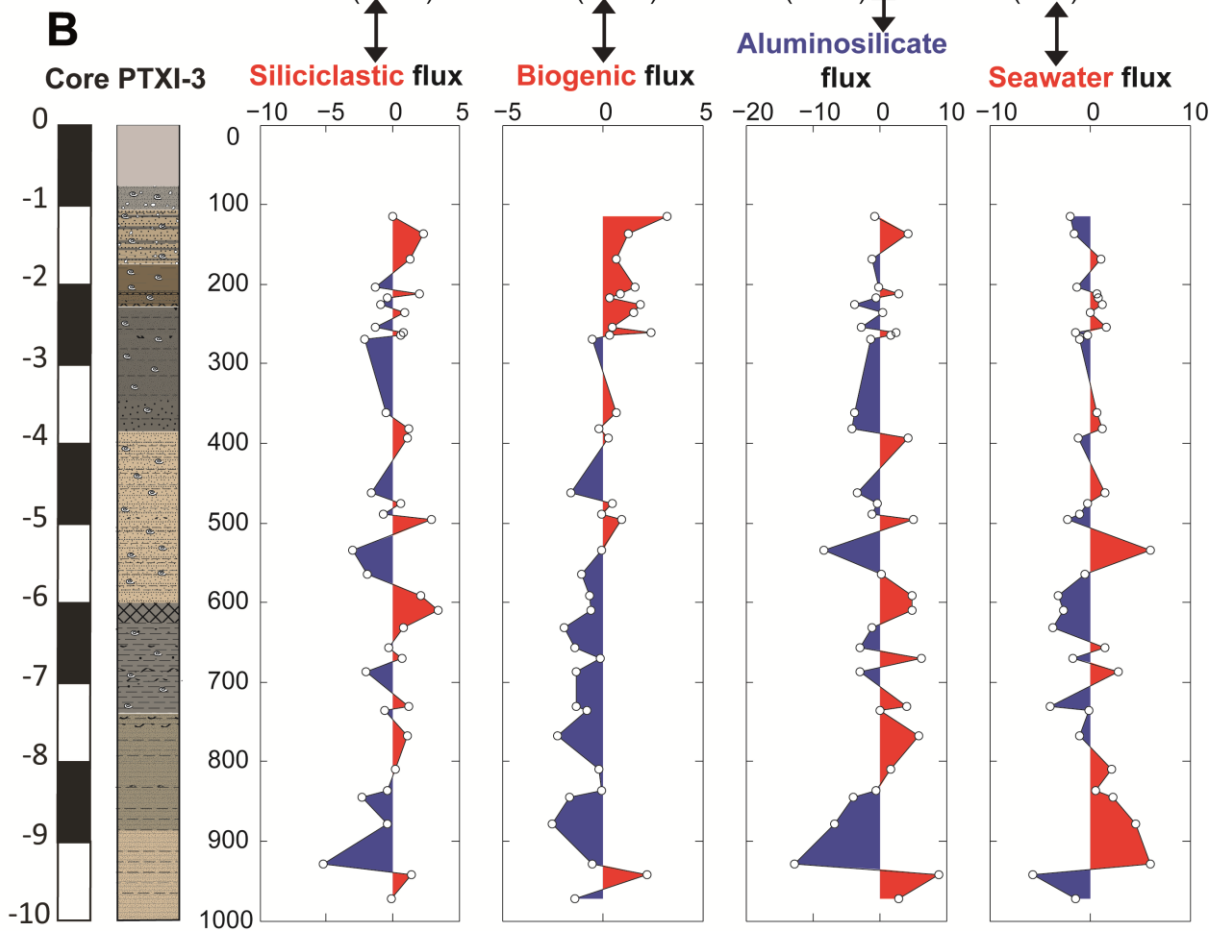
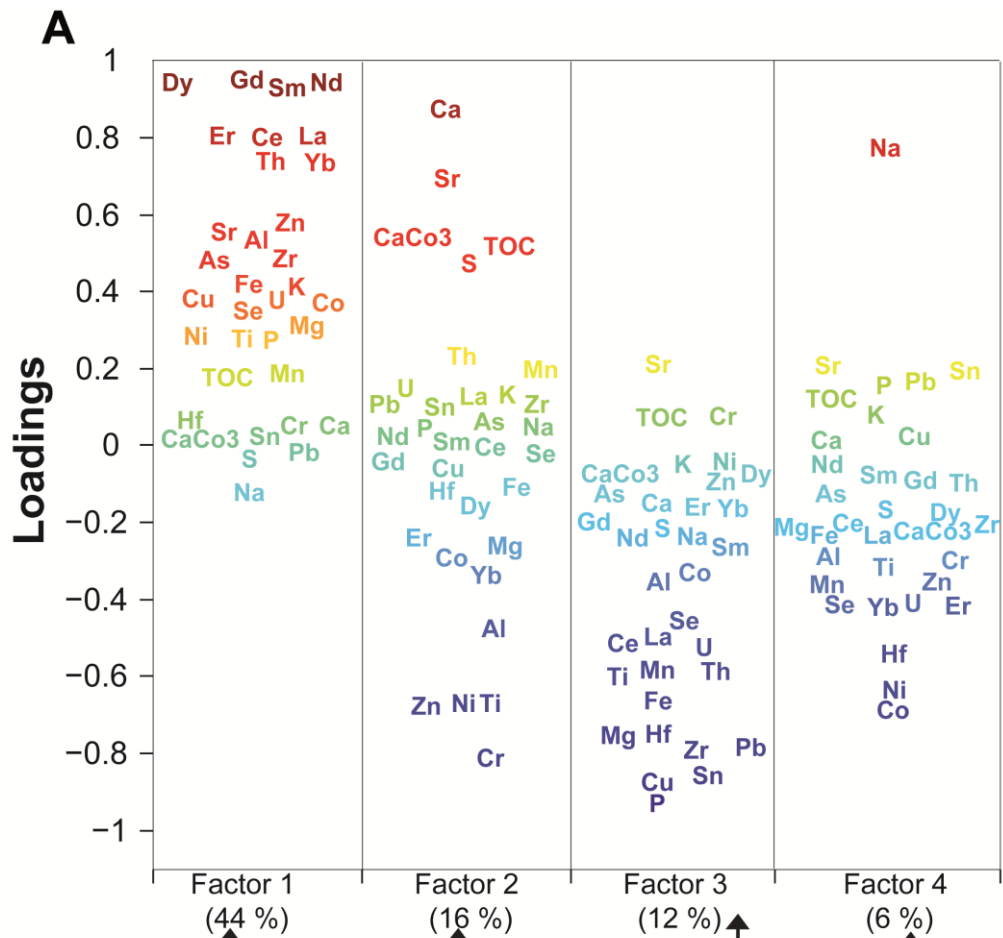
1186 pluricentimetric layer containing placers.



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1188 **Figure S4.** (A) Factor analysis of elemental concentrations in core PTXI-3 taken in the
1189 ancient harbor basin of Claudius. The number of factors is limited to four: (1) Silicate detrital

- 1190 fraction (44%); (2) biogenic fraction (16%); (3) aluminosilicate detrital fraction (12%); (4)
- 1191 seawater influence (6%). **(B)** Distribution of the different factors with depth in the column.



1193 **Table S1.** Major and trace element concentrations in core PTXI-3. Values in wt.% for major
1194 elements and in ppm for trace elements.

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