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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

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

basins. From this comparative approach, it appears that the respective weight of each control 25 factor is dependent on the coastal geomorphological context of the sites where the harbors 26 were established. Since the discovery of the harbor of Ostia Antica in 2014, this method has 27 provided the means, for the first time, to identify two distinct harbor basin regimes; an initial 28 marine-dominated regime from the middle of the 4th c. BC to ~ the 3rd c. BC, and a later 29 freshwater-dominated regime up to the 2nd c. BC. More generally, we observe the effects of 30 the dynamics of the deltaic progradation of the Tiber, which very early on was subject to a 31 hydro-climatic component, on the processes of alluviation of the harbor basin. Additionally, 32 and also for the first time in harbor geoarcheology, Pb isotope compositions measured 33 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 particles. The present study further provides an opportunity to test the validity of two 36 hypotheses recently put forward: (1) that a series of three tsunamis is recorded in the harbor 37 silts, and (2) that an initial lagoon-type harbor was constructed at Ostia Antica, which later 38 evolved into a fluvial harbor. Neither of these hypotheses are supported by the present data. 39 40 Keywords: Rome, Ostia Antica, ancient harbor basins, geoarcheology, sedimentology, 41 42 geochemistry, Pb isotopes, Tiber river 43 1. Introduction 44 45 When harbor geoarcheology emerged as a discipline at the beginning of the 1990s following the excavations of the ancient harbors of *Caesarea* (Israel) (Reinhardt et al., 1994, 46

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;

Bravard, 2009; Cubizolle, 2009). Two decades later, while these well-tested methods continue
to be used in the field, elemental and isotopic geochemistry has been added successfully to the
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 al., 2015b), and Alkinoos (Finkler et al., 2018)

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

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

racing the such common mistake consists in using certain elemental ratios capable of tracing the

real sedimentological (e.g. Zr/Rb and Zr/Ti applied as particle size proxies) or environmental (e.g.

Sr/Al, Mg/Ca or Ca/Al, and Si applied as biological production proxies or Mn/Fe, U, and Mo 75 applied as water ventilation proxies) parameters without taking into account the geological. 76 77 hydrological, and climate contexts of the sites under study, each of which can significantly modify the meaning of these ratios. Initially, the use of geochemistry for reconstructing 78 environmental processes was applied to (i) marine sediments to decipher paleo-productivity, 79 deep water ventilation, and paleo-temperatures, and (ii) lacustrine sediments to track 80 atmospheric pollution, human impact, paleo-climatic changes, aeolian input, paleo-floods, 81 catchment weathering, and source rock compositions (Boyle, 2002; Croudace and Rothwell, 82 2015). 83 84 Here, we discuss the types of environmental and anthropogenic processes that are most

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

(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

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.

Improving this understanding is all the more important since Ostia Antica had a river harbor located a short distance from the mouth of the Tiber River, which was to subject it to both marine and river influences (Goiran et al., 2012, 2014, 2017; Salomon et al., 2016). As a result, in the present work, we focus in particular on the interaction between fluvial and 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 Marcius in the 7th c. BC. However, the *castrum* (building, or plot of land, used as a fortified 108 military camp) did not appear before the 4th or beginning of the 3rd c. BC (Coarelli, 1988; 109 Martin, 1996; Zevi, 2002; Brandt, 2002). The absence of evidence for the earliest periods has 110 led to hypotheses about the existence of a paleo-Ostia, the location of which remains 111 unknown and would correspond to the city founded by Ancus Marcius (Coarelli 1988; Martin 112 113 1996; Zevi 2002; Brandt 2002). Ancient texts reveal that the harbor of Ostia Antica became a commercial and military port during the Punic wars (Livy, 45, 22) and played a vital role in 114 providing Rome with food supplies (especially olive oil from Spain and grain from the North 115 African provinces) during the 1st c. BC (Le Gall, 1953; Zevi, 2001b). Nevertheless, at this 116 time, Puteoli (Pozzuoli) on the Bay of Naples remained the principal maritime port of Rome 117 for trade (Balland, 1965). It was considered to be one of the busiest centers of trade in the 118 Roman Empire, especially for metal ingots, for which a route from the Cartagena/Mazarron 119 and Rio Tinto mines in Spain to Puteoli and Rome was heavily used (Domergue and Rico, 120 121 2014; Delile et al., 2016b). The large distance separating the ports of Puteoli and Ostia Antica, as well as a problematic massive siltation of Ostia's harbor basin caused by a succession of 122 floods discharging sediments at the mouth of the Tiber (Strabo, 5, 3, 5), combined with the 123 need to accommodate an increasing number of merchant and military ships arriving from the 124

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

128 2.2. Geography and geology

The Tiber delta constitutes the outlet for the water and sediment discharges of a river 129 130 405 km long which drains a watershed surface area of 17,375 km², comprising young sediments from the Apennines and modern volcanic deposits in Latium. The annual 131 deposition of sediment in the Tyrrhenian Sea, estimated at 7.2 million tons per year (Iadanza 132 133 and Napolitani, 2006), has resulted in the construction of a wave-dominated delta 150 km² in extent. The Tiber delta can be divided into (i) the eastern inner deltaic plain, related to the 134 paleo-lagoons of Maccarese in the north and Ostia in the south; (ii) the western outer deltaic 135 plain, where the landscape is composed of accreting dunes to which must be added the 136 submerged area; (iii) the deltaic front (mainly composed of sand and silt); and (iv) the 137 prodelta (mainly composed of mud and clay) (Salomon, 2013; Salomon et al., 2017). 138 The geomorphological evolution of the Tiber delta during the Holocene period is well 139 documented (Bellotti et al. 1994, 1995, 2007, 2011; Giraudi, 2004; Milli et al., 2013; 140 141 Salomon, 2013; Salomon et al., 2018). During the periods of the Republic and the Empire, the position of the Tiber mouth likely was located close to the Boacciana Tower (Tomassetti 142 **1897**) (Fig.1B). Additional information about the coastline comes from the immediate 143 surroundings of *Portus* (Arnoldus Huyzendveld, 2005; Giraudi, 2004; 2009; Salomon, 2013) 144 and from south of the Tiber delta, on the Laurentine shore (Bicket et al., 2009). However, 145 during the Imperial period, the rapid seaward progression of the coastline recorded by the 146 Tiber delta, which could have been more active at the river mouth, is the cause of relatively 147 poor paleo-geographical knowledge of the Tiber mouth and its dynamics during Roman times. 148 This rapid evolution explains why discovering the exact location of the ancient port of Ostia 149

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

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

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

We analyzed at high resolution one (PO2, Fig. 1B) of the two cores drilled by Goiran et
al. (2014) which led to the discovery of the first harbor of Rome at Ostia Antica. This 12
meter long core was drilled in a low-lying area without any archeological structural remains
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-

depth model and the ¹⁴C dates, as well as the inherent uncertainties associated with both, can

be found in Delile et al. (2017), who exercised great caution in the use of these ages by

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

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

The geochemical analyses were carried out on the entire core with high sample resolution (86 samples analyzed, i.e., one sample approximately every 14 cm). The data are published in Delile et al. (2017) together with the analytical techniques, which are also briefly summarized here. First, the sediments were sieved at 63 µm with a nylon sieve instead of a 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 clean laboratory under laminar flow hoods. The samples were digested in a 3:1:0.5 mixture of 199 concentrated double-distilled HF, HNO₃, and HClO₄ in Savillex beakers left closed on a 200 hotplate at 120-130 °C for 48 h, then evaporated to dryness. Perchlorates and any remaining 201 fluorides were converted to chlorides by drying down several times with 6 M distilled HCl. 202 The samples in solution in 6 M distilled HCl were all clear, indicating complete sample 203 breakdown. The samples were re-dissolved in 2 ml concentrated distilled HNO₃, from which 204 ~10 percent aliquots were further diluted to 2% HNO₃ and internal standards (10 ppm Sc for 205 ICP-AES and 2 ppb In for Q-ICP-MS) added. Major elements were analyzed by ICP-AES 206 207 (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 elements. The precision on major and trace elements is between 2 and 5%. 209

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

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

232

4. Results and discussion

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

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

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

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

(i) Detrital material (Fig. 3A) is derived from erosion of continental crust and volcanic 247 rocks. These erosion products are carried by the hydrological network from their sources at 248 the watershed to the marine environment. The principal elemental components of the particle 249 flux transported by modern fluviatile sources are, in order of decreasing abundance, Si, Al, 250 Fe, Ca, K, Mg, Na, Ti, P, Mn, and Ba, according to Martin and Whitfield, 1983. However, 251 inter-comparison of these elements within the PO2 core shows stratigraphic distributions (see 252 253 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 relatively weak levels of correlation between, for example, Al and K (r = 0.31), Mg and Fe (r255 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 observations demonstrate that it is incautious to use *sensu stricto* certain elements considered 258 259 to be universal markers of detrital origin. These differences between observation and theory in the chemical composition of the detrital fraction are accounted for by the relative weight of 260 the local specificities that directly affect the factors controlling the composition in question: 261 the mineralogy of the enclosing rocks, the erosional regime which itself depends on regional 262 263 climate conditions (temperature, precipitation, and runoff), and the reactivity of eroded 264 material during transport (Sageman and Lyon, 2003).

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

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

(iii) *Biogenic material* (Fig. 3C) reflects the photosynthetic production of organic 281 material and the presence of remains of skeletal material, the latter being composed of calcite 282 (CaCO₃) or silica (SiO₂). Biogenic fluxes in the sedimentary system depend on the 283 concentration and supply of nutriments from surface waters (nitrate, phosphate, and micro-284 nutriments such as Fe). They are produced by chemical erosion and terrestrial biogenic 285 286 activity or biological processes in the water column. Usually, the biogenic flux is tracked by Si, Ca, Sr, P, or Loss on Ignition (L.O.I.) (Sagement and Lyons, 2003; Sánchez Vizcaíno and 287 Cañabate, 1999; Delile et al., 2016a). However, as observed for the detrital and authigenic 288 fluxes, neither the qualitative nor the quantitative observations (Figs. 3C and S1) show clear 289 systematic positive or negative correlations between, for example, Ca and Sr (r = 0.04), Ca 290 and P (r = -0.3), and Sr and P (r = 0.3). 291

This attempt at classification, or grouping, of chemical elements taken individually according to the three principal inputs involved in the formation of sedimentary deposits (Fig. 3) has revealed the limits of the approach. First, assigning a given element to a specific category of inputs is not supported by robust statistic correlations, hence calling for caution in automatically using geochemical proxies traditionally considered to be robust tracers for particular inputs. Second, the individual observation of several dozen chemical elements is
fastidious work without any real added value. For these reasons, the reading of the underlying
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

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

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

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

Roman Magmatic Province – Latian District (RMP-LD) (Mattei et al., 2010) which regroups 321 the Colli Albani, Vulsini, Cimini, Vicano, Ceriti, and Sabatini volcanoes, all very close to the 322 Tiber delta. The RMP-LD region, which is one of four major volcanic provinces of the 323 Tyrrhenian margins of peninsular Italy, consists of mafic ultrapotassic rocks with a 324 remarkably homogeneous elemental composition characterized by high abundances of Pb, Th, 325 U, Ce, and La and low abundances of Ti, P, Ta, and Nb (Mattei et al., 2010). These 326 geochemical features distinguish all the volcanoes close to the Tiber delta. This is shown in 327 Fig. 4A where elements with affinities for the RMP-LD mafic ultrapotassic rocks show 328 strongly negative values (ranging between -0.7 and -1), whereas elements depleted in the 329 330 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 particles, since the lowest F1 values occur in the sandy units A and B2 (Figs 2, 4B). The 332 correlogram for elementary and granulometry data (Fig. S2) from harbor units only (units B1 333 and B2) shows that the sandy fraction is clustered within these elements (Th, Zr, U, K, REEs, 334 etc.). We conclude from this that Factor 1 reflects siliciclastic terrigenous influence which, in 335 terms of depositional environment, corresponds to the ancient hydrodynamic conditions of the 336 harbor water column. 337

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

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

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

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

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

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

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

analyze navigation as it reduces the draught for ships in the harbor basins because of siltation and

alluviation (Goiran and Morhange, 2003; Marriner and Morhange, 2007; Salomon et al.,

2016; Goiran et al., 2017). The predominance of detrital fluxes in the paleo-environmental 398 evolution of the water column in harbor basins is subject to the coastal geomorphology of the 399 sites where these harbors were established, as only those located at deltas are dominated by 400 this environmental parameter. Indeed, the paleo-environmental changes of the water column 401 of the ancient harbor of Naples, located in a small coastal bay, were affected more by aeration 402 (Factor 1 = redox conditions = 36%) than by terrigenous contributions (Factor 2 = 16%) (see 403 Table 1 and/or Delile et al., 2016a). In general, these detrital fluxes are composed of 404 405 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 hydrodynamics of harbor basins in which the recrudescence of currents is produced either by 407 408 the activity of canals connecting the harbor basins to the river (in freshwater-dominated harbor environments), or by the acceleration of deltaic progradation (on floodplains, coastal 409 alluvial fans, paleo-channels, delta-fronts, and prodeltas) (Table 1). 410

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

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

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

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

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

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

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

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

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

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

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

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

modify the mineralogy and thus the geochemistry of the transported sediments. According to 518 Garçon et al. (2014), a "heavy mineral effect" alters the Pb isotopic budget of both coarse and 519 fine fluvial sediments. Some heavy minerals concentrated in coarse sediments are extremely 520 radiogenic, allowing sandy sediments to grow in more radiogenic ²⁰⁸Pb/²⁰⁶Pb values (Garcon 521 et al., 2014) and, thereby, acquire elevated values of κ (r between ²⁰⁸Pb/²⁰⁶Pb vs $\kappa \sim 0.7$). The 522 sandy deposits of the Tiber delta's littoral facies are strongly enriched in heavy minerals 523 (Belfiore et al., 1987) that take the form of placers (Salomon, 2013) (Fig. S3) derived from 524 the erosion of the Monti Sabatini or the Colli Albani (Funiciello, 1995; Ventriglia, 1971). The 525 presence of such minerals requires a source close to the deposit area (Carling and Breakspear, 526 527 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 $f\alpha$ ' measured in core PO2 (r = -0.7).

533 *4.4. Chronology of the life cycle of the harbor*

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

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

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

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

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

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

Gap 3 is identified by a sharp drop in the sedimentation rate between the last date of unit B1 at ~ 585 cm depth (387-204 BC) and the first date from unit B2 at 526 cm depth (163 BC-AD 16). This chronological gap reflects a second potential dredging phase that took place at the earliest before the mid- 2^{nd} c. BC. Gap 4 is identified by a last sharp drop in the sedimentation rate between the date at 378 cm
depth (158 BC-AD 24) and that at ~ 550 cm depth (AD 80-231). This chronological gap
reflects a third and last potential dredging phase that took place at the latest during the 1st c.
AD.

570 *4.4.2. The pre-harbor environment*

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

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

595 *4.4.3. The lower harbor sequence*

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

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
and 3.8 m *b.a.s.l.*) reflects a marine-dominated regime since the F4 values (seawater
influence) always remain positive, although being lower than those of unit A due to the
connection of the harbor basin to the Tiber river;

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
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 this616 subunit (Fig. 2).

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

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

639 *4.4.4. The upper harbor sequence*

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

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

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

(i) During the time period covered by unit B2 (2^{nd} c. BC to 3^{rd} c. AD), a major hydrosedimentary event, which is characterized by an increase in the frequency of floods, occurred during the period of the 1^{st} c. BC to the 2^{nd} c. AD approximately at several places in the western Mediterranean region (Berger and Bravard, 2012). Based on the flood events of the Tiber River in Rome recorded by Bersani and Bencivenga (2001), corroborating the work of Le Gall (1953), such an increase in the flood frequency of the Tiber took place between approximately the 2^{nd} c. BC and the 2^{nd} c. AD.

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

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

706 *4.4.5. The abandonment of the harbor basin*

The yellow bedded silts of unit C (Figs. 2 and 4) from the 3rd c. AD onward represent floodplain deposits which definitively isolated the harbor basin from the sea. Unit C presents the same geochemical characteristics as the floodplain deposits recorded in the uppermost unit (unit C) of the Trajan basin at *Portus* (Delile et al., 2014a). In both cases, the depositional environment was freshwater (negative values of F4), oxygenated conditions prevailed (constant positive values of F3), and the detrital particles dominated by the aluminosilicate minerals demonstrate the fineness of these sediments (high positive values of F2 and F1) (Fig. 4). These characteristics indicate that the abandoned harbor had finally been overrun by thefloodplain around five centuries before the harbor of Trajan suffered the same fate.

- This abandonment phase of the harbor basin appears to have been mistaken with the 716 717 establishment of a fluvial harbor (Hadler et al., 2015) called the "younger fluvial harbour phase" in Wunderlich et al. (2018). Indeed, these authors argue for a shift from a harbor of 718 lagoon type towards a fluvial harbor phase (or "harbour generation II") located between - 0.5 719 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 + 720 0.3 m above a.s.l. These values, inferred from the ancient biological sea level dated to AD 721 230-450 by Goiran et al. (2009), show unambiguously that the water column disappears at 722 723 these depths because the environment became terrestrial. Consequently, the establishment of a river harbor at these depths is strictly impossible. For this reason, we strongly suggest using 724 725 ancient sea levels to characterize harbor levels and phases rather than the simple measurement of present sea level. 726
- 727

728 5. Conclusions

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

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

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

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

and the 3rd c. AD because the harbor basin was affected by the high-energy floods of the Tiber 763 764 river. During this period, the geochemical signal of the coarse harbor deposits carefully recorded the hydrological rhythms of the Tiber, marked by the alternation of major inundation 765 phases and pronounced low-water phases. Again during this period a phase of increased 766 progradation of the Tiber delta occurred, resulting from an increase in the flood frequency of 767 the Tiber between approximately the 2^{nd} c. BC and the 2^{nd} c. AD. While Hadler et al. (2015a) 768 and Wunderlich et al. (2018) have advocated for a high-energy event to interpret the coarsest 769 B2 unit, our geochemical data are more consistent with the scenario proposed by Goiran et al. 770 (2014), Sadori et al. (2016) and Salomon et al. (2016) who call on an upsurge in the frequency 771 772 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 environment, as argued by Hadler et al. (2015a) and Wunderlich et al. (2018), since these 774 775 deposits are located in part above ancient sea level (-0.2 to +0.3 m *b.a.s.l.*); the harbor water column no longer existed at that time. 776

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

deposits. However, our multi-elemental geochemical signal records the exact opposite, a

strong freshwater component that increases towards the top of the stratigraphy.

789

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- 1128 Figure 1. (A) Location of the harbor basins of *Portus* and Ostia in the Tiber delta and position
- 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.



Figure 2. Stratigraphic log of core PO2 showing the age-depth model, stratigraphic and
environmental descriptions, magnetic susceptibility values, the 50 percentile (D 50) grain

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



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









- 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).



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



- **Table 1.** Environmental factors involved in the formation of sedimentary deposits in the
- ancient harbor basins of Ostia (Factor analysis in Fig. 4), Claudius (Factor analysis in Fig. S4
- and data in Table S1), Trajan, Ephesus, and Naples. These environmental variables, organized
- 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%)					
		Cignificant alamanta	Al, Zr, Ti, Th, Mg, Ba, K, Co,	LOI, Ca, Sr	Mn, K, Ti, Zn Cr, Co, Cd	S, Mo, U, Pb, Ag, Cu	Al, Rb, La, Se, Ce, Ga, Li, Be,	Pb, Ag, Sn, Cu, Cd,	Na					
		Significant elements	Zn, REEs				Zr, K, Ti, Mg							
			Fluvial hydrodynamics:	Biomass production:	- Delta progradation	-Enclosing harbor basin	- Erosion of the local and	- Heavy metal inputs	- Seawater influence					
			 Canal networks 	 Organic matter 	- Swell	-Weak water-column	regional geological	 Using of lead pipe 	 Infiltration of the 					
		Drocoss	 Coastal and delta 	 Skeletal remains 		ventillation	basement	networks	littoral salt wedge					
		FIDCESS	progradation	 Eutrophication 		-Interflood period		 Metallurgic 						
				 Urban sewage inputs 		-Urban sewage inputs		activities						
SVNTU						and eutrophication								
51111	1313		- Floodplain	- Swamp	- Floodplain	- Marine			- Marine dominated					
			- Freshwater-dominated	- Marine	- Freshwater-dominated harbor	- Enclosed harbor			harbor					
			harbor	- Enclosed harbor	- Coastal alluvial fan	- Peat			- Delta front					
			 Coastal alluvial fan 	- Peat	- Paleo-channel	- Lagoonal	In all any incompanie	In all any drammants						
		Paleo-environment	- Paleo-channel	- Lagoonal	- Delta front	- Swamp	in an environments	in all environments						
			- Delta front	- Brackish and eutrophic	- Prodelta environments									
			- Prodelta environment	environments	- Beach									

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



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



Figure S3. (A) Photo showing subhorizontal bedding formed by placers on the current
coastline of the Tiber delta (modified from Salomon, 2013). (B) Close-up of a

1186 pluricentimetric layer containing placers.



Figure S4. (A) Factor analysis of elemental concentrations in core PTXI-3 taken in the

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)
- seawater influence (6%). (**B**) Distribution of the different factors with depth in the column.



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

			wt.%										ppm																					
Depth (cm)	L.O.I.	CaCo3	Al_2O_3	Fe ₂ O ₃	CaO	Na ₂ O	K ₂ O	MgO	TiO ₂	P_2O_5	MnO	SO ₂	Zr	Cr	Со	Ni	Cu	Zn	As	Se	Sr	Sn	La	Ce	Nd	Sm	Gd	Dy	Er	Yb H	lf	Pb	Th	U
115	5	33	8.76	3.37	19.66	1.28	2.15	1.49	0.30	0.14	0.10	0.39	146.16	34.88	13.64	28.65	25.72	41.07	28.02	9.86	652.26	2.94	57.31	111.69	49.62	8.62	6.91	4.48	2.30	1.92 16	1.11	44.12	25.89	3.16
136	6	40	7.84	2.63	20.67	1.42	2.09	1.37	0.24	0.10	0.12	0.41	121.39	35.68	12.24	31.88	25.10	52.89	22.17	3.22	981.79	2.73	52.05	100.85	54.40	9.88	7.41	5.23	2.43	1.88 59	.21	47.06	23.72	3.29
169	30	13	8.25	2.36	17.88	1.46	2.20	1.26	0.27	0.11	0.12	0.78	134.73	38.27	7.11	23.51	29.19	57.61	17.48	4.69	1037.52	3.81	56.46	112.99	61.41	10.94	8.65	6.00	2.76	2.42 63	.92	55.27	26.82	4.38
205	8	26	6.26	3.93	18.14	1.01	1.51	0.91	0.22	0.09	0.10	11.14	139.48	3 29.97	9.63	22.03	20.40	35.09	19.90	7.80	577.20	1.07	36.17	70.02	32.11	5.92	4.83	3.45	1.81	1.59 12	7.19	38.09	16.41	2.68
213	14	23	9.08	3.80	16.51	1.53	2.45	1.03	0.23	0.10	0.07	0.84	84.42	37.02	9.39	26.95	16.23	54.87	23.13	5.29	908.35	3.08	48.67	95.10	51.77	8.68	6.98	4.85	2.10	1.82 46	.36	35.02	22.59	3.51
218	16	26	8.28	4.20	18.97	1.35	2.28	0.99	0.22	0.10	0.07	3.10	97.62	26.88	7.82	19.98	18.68	46.45	25.03	3.47	902.39	2.64	45.63	87.70	47.59	7.84	6.41	4.04	1.89	1.62 45	.28	34.33	20.79	3.45
226	23	15	8.50	5.12	14.76	1.53	2.11	1.18	0.25	0.11	0.11	3.92	138.14	43.83	7.86	28.31	21.37	56.93	19.39	3.59	827.42	2.70	44.63	86.17	47.76	8.41	6.46	4.43	2.05	1.77 58	.44	41.19	23.73	3.77
235	17	27	8.81	2.99	18.65	1.41	2.30	1.24	0.25	0.10	0.06	2.30	88.53	42.18	9.92	32.18	18.00	61.85	21.94	7.95	1073.60	3.31	51.25	98.90	55.19	9.53	7.46	4.87	2.34	1.85 53	.49	38.47	23.36	3.74
255	2	34	8.40	2.65	17.00	1.49	2.37	1.06	0.23	0.10	0.06	1.74	88.06	27.78	8.53	19.94	15.64	44.85	20.43	3.90	842.42	4.40	41.52	81.16	42.96	7.73	6.35	4.04	1.82	1.55 44	.53	39.55	18.95	6.41
262	11	28	9.15	3.46	17.78	1.40	2.32	1.31	0.30	0.11	0.14	2.08	111.33	34.77	13.28	25.11	13.54	40.69	19.01	6.56	609.94	1.18	45.82	87.19	40.23	7.02	5.42	3.84	1.83	1.52 13	7.32	36.62	21.38	8.71
265	4	33	8.74	3.40	15.39	1.45	2.24	1.25	0.27	0.10	0.06	2.65	97.61	43.23	9.17	25.41	19.58	56.97	18.58	5.96	758.57	3.59	43.56	83.38	48.71	8.19	6.38	4.40	2.11	1.80 53	.04	37.13	19.52	4.98
270	13	26	8.35	2.92	17.10	1.45	2.29	1.18	0.26	0.11	0.12	1.89	98.31	28.42	10.42	19.22	11.40	34.98	12.76	6.90	567.53	1.48	37.20	73.88	32.21	5.86	4.71	3.40	1.61	1.25 11	0.13	36.47	18.09	5.51
362	7	25	9.19	5.77	19.91	1.58	1.63	2.14	0.48	0.27	0.14	3.98	204.82	41.62	12.54	24.44	73.43	54.09	16.69	8.05	571.24	111.75	60.79	124.43	53.74	9.44	6.97	4.33	2.00	1.71 21	5.25 2	2100.79	30.22	6.30
382	4	23	10.90	6.16	16.28	1.73	2.63	3.11	0.55	0.35	0.16	2.08	274.70	42.85	16.58	29.13	117.46	65.84	25.63	11.75	678.14	47.82	77.05	159.96	66.02	11.72	8.67	5.22	2.56	2.12 28	6.91 5	562.95	43.26	8.39
393	10	25	7.61	2.96	17.83	1.24	1.88	1.54	0.33	0.10	0.05	0.33	74.98	85.89	11.18	35.67	16.34	57.35	7.33	9.91	709.48	2.40	36.13	79.71	42.45	8.11	6.26	4.40	2.07	1.68 49	.58	28.58	12.55	2.59
463	6	34	7.23	2.95	15.83	1.23	1.76	1.83	0.34	0.11	0.05	0.44	87.22	93.57	17.94	40.24	19.85	63.36	9.17	1.51	651.93	1.94	35.48	81.67	46.99	8.29	6.61	4.38	2.08	1.69 56	.01	30.16	11.97	2.59
475	3	25	8.57	3.06	14.67	1.34	1.93	1.40	0.35	0.11	0.09	0.23	99.40	56.60	11.29	36.73	18.95	63.35	7.64	7.06	709.13	3.76	49.74	110.23	53.51	9.06	7.71	5.17	2.39	2.03 59	.45	39.51	19.08	5.03
490	1	21	8.14	3.26	16.03	1.26	1.79	1.39	0.35	0.10	0.11	0.09	108.00	74.28	11.95	42.11	16.74	58.20	8.62	7.43	608.66	0.77	49.15	106.25	46.55	8.38	6.56	4.43	2.37	1.95 203	2.93	37.29	18.89	5.80
496	1	21	9.25	3.51	14.92	1.36	2.12	1.42	0.36	0.12	0.10	0.28	122.63	54.55	11.70	30.04	21.84	71.99	8.41	7.92	887.80	2.63	52.83	115.08	58.60	10.62	7.92	5.20	2.17	1.83 65	.20	47.48	22.51	3.79
535	1	21	7.67	2.82	13.50	1.37	1.81	1.45	0.34	0.10	0.09	0.19	90.28	67.39	9.22	28.28	16.50	54.45	5.26	3.22	566.48	2.14	31.73	72.48	39.94	7.49	5.87	4.16	1.92	1.58 47	.67	27.62	12.08	2.50
564	1	14	8.09	2.79	16.58	1.41	2.23	1.23	0.25	0.08	0.05	0.23	73.16	47.03	9.18	27.49	13.05	44.91	4.79	2.87	606.56	1.24	26.93	58.22	30.93	5.81	4.49	3.43	1.47	1.26 35	.08	25.98	9.37	1.55
591	1	19	8.49	3.59	15.02	1.27	1.92	1.54	0.42	0.12	0.06	0.33	111.94	116.68	11.88	39.37	22.06	76.78	8.77	7.67	682.82	3.16	50.57	114.34	59.54	10.22	7.72	5.19	2.40	1.96 70	.02	41.09	21.14	3.32
611	1	13	9.95	5.02	10.09	1.37	2.00	1.24	0.37	0.12	0.04	4.00	141.79	55.51	14.62	44.00	33.03	75.24	52.61	8.64	621.30	3.19	65.71	133.91	71.13	12.07	9.44	6.12	2.73	2.23 70	.37	61.35	27.14	4.30
633	1	19	13.56	5.38	8.47	0.93	2.23	2.17	0.62	0.11	0.13	0.30	171.31	119.39	33.17	94.70	46.05	125.25	19.62	10.58	345.19	4.01	65.24	131.49	58.35	10.39	8.31	5.92	3.18	2.90 28	9.57	48.00	26.87	9.51
657	2	11	10.17	4.45	10.50	1.31	2.17	1.52	0.40	0.11	0.12	1.40	139.98	86.28	14.14	45.09	31.44	80.76	67.09	4.95	559.47	3.55	45.11	99.90	49.55	8.72	7.13	5.39	2.47	2.15 66	.00	43.10	19.86	4.05
670	10	10	7.91	2.28	12.03	1.50	2.05	1.07	0.26	0.08	0.04	0.21	62.44	64.37	7.08	24.31	13.92	45.46	7.40	2.80	499.08	2.06	25.22	58.02	29.82	5.09	4.31	3.08	1.45	1.22 35	.67	24.76	10.13	1.92
688	5	12	7.99	2.53	10.88	1.53	1.90	1.17	0.32	0.09	0.09	0.15	65.11	54.42	6.75	35.52	13.64	49.49	9.80	8.01	408.44	1.11	34.54	75.85	34.08	6.28	5.32	3.76	1.86	1.69 11	7.41	26.90	10.25	2.14
731	5	18	11.32	4.84	10.23	1.30	2.13	2.06	0.50	0.11	0.13	0.28	110.78	8 102.58	12.87	61.21	29.09	85.63	17.70	7.60	346.67	2.14	42.14	87.74	39.62	7.26	6.04	4.57	2.54	2.21 17	7.41	35.79	17.28	3.30
736	1	14	9.15	3.39	11.41	1.45	1.96	1.36	0.40	0.12	0.05	0.28	99.40	71.20	8.37	32.28	20.51	63.49	15.35	4.52	476.60	2.19	35.98	82.03	42.00	7.73	5.82	4.16	2.04	1.92 57	.94	32.65	15.97	2.40
768	8	22	8.55	3.07	11.40	1.55	1.93	1.22	0.33	0.11	0.09	0.15	96.40	61.84	8.28	25.69	18.27	60.80	7.65	4.55	477.03	1.66	31.44	69.01	38.18	6.92	6.04	4.21	2.06	1.80 48	.59	27.54	14.64	2.37
809	2	12	8.84	2.90	11.95	1.52	2.13	1.11	0.28	0.10	0.04	0.14	75.35	67.83	8.55	30.78	18.10	55.69	8.73	2.06	504.85	1.55	31.30	68.00	37.41	6.55	5.56	3.81	1.79	1.52 43	.48	30.15	13.48	1.96
837	1	13	8.57	2.98	11.74	1.60	1.88	1.25	0.35	0.12	0.05	0.16	77.27	75.11	9.39	31.38	18.24	61.56	7.74	2.47	446.30	2.26	31.64	68.22	38.17	6.54	5.77	4.07	1.93	1.69 49	.22	24.16	12.87	2.49
845	2	20	8.22	2.93	11.88	1.54	1.91	1.19	0.38	0.10	0.09	0.05	80.95	71.42	8.18	33.82	12.29	49.10	9.98	6.23	449.30	1.55	33.30	71.41	34.79	6.19	5.03	3.70	1.98	1.68 13	6.89	25.80	12.58	2.31
879	1	17	11.82	4.36	12.35	1.28	2.29	1.36	0.43	0.13	0.05	0.16	132.24	69.86	12.26	44.82	29.43	89.69	18.60	8.06	609.53	2.59	61.77	125.12	63.32	10.71	8.06	5.39	2.40	2.11 74	.81	61.45	29.79	3.34
928	3	22	7.89	2.48	13.56	1.51	2.04	1.08	0.28	0.08	0.04	0.14	54.95	68.45	8.16	29.54	13.48	52.44	7.47	6.35	553.82	1.41	30.63	65.05	35.85	6.54	5.43	3.87	1.82	1.46 37	.72	22.84	10.68	1.72
942	1	21	7.52	2.25	12.65	1.45	2.01	1.00	0.25	0.08	0.04	0.13	57.06	52.04	6.92	23.65	12.74	47.00	6.95	1.67	506.07	1.04	23.20	49.78	28.72	5.35	4.52	3.29	1.54	1.35 35	.31	20.16	8.46	1.40
973	1	22	7.63	2.40	11.86	1.55	1.76	1.09	0.36	0.09	0.08	0.07	75.65	60.11	7.60	45.66	10.49	50.08	10.07	5.39	405.26	0.64	34.49	70.03	33.34	6.40	5.51	3.87	1.98	1.81 12	2.32	21.42	11.12	2.45