Demise of a harbor: A geochemical chronicle from Ephesus 1 2 3 Authors version of an article in JOURNAL OF ARCHAEOLOGICAL SCIENCE · OCTOBER 2014 DOI: 10.1016/j.jas.2014.10.002 4 Hugo Delile^{1,2*}, Janne Blichert-Toft^{2,3}, Jean-Philippe Goiran⁴, Friederike Stock⁵, Florent 5 Arnaud-Godet², Jean-Paul Bravard¹, Helmut Brückner⁵, Francis Albarède^{2,3}, 6 7 8 ¹Université Lumière Lyon 2, CNRS UMR 5600, 69676 Bron, France 9 ²Ecole Normale Supérieure de Lyon, Université Claude Bernard-Lyon 1, CNRS UMR 5276, 69364 Lyon Cedex 10 7. France ³Department of Earth Science, Rice University, Houston, TX 77005, USA 11 ⁴Maison de l'Orient et de la Méditerranée, CNRS UMR 5133, 69365 Lyon Cedex 7, France 12 13 ⁵Institute of Geography, University of Cologne, Albertus-Magnus-Platz, 50923 Cologne (Köln), Germany 14

- 15 *Corresponding author: <u>hdelile@gmail.com</u>Tel: +33 6 82 73 66 53
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17 Abstract

At the end of the first century BC, Ephesus became the Roman capital of Asia Minor and the 18 most important commercial, religious, and cultural center of the region. In order to evaluate 19 the status of anthropogenic fluxes in the port of Ephesus, a 12 m long sediment core drilled in 20 the Roman basin was investigated to shed light on the paleo-environmental evolution of the 21 harbor using grain size distribution analysis, ¹⁴C ages, major and trace element geochemistry, 22 and Pb isotope compositions. With the help of complementary sedimentological data and 23 Principal Component Analysis, five distinct units were identified which, together, reflect the 24 25 different stages of water history in the harbor. Among the major disruptive events affecting the port were earthquakes and military events, both of which were particularly effective at 26 destroying the water distribution system. 27

Seasonal floods of the Cayster River (Küçük Menderes) were the major source of the silt that progressively infilled the harbor. Silting in was further enhanced by the westward migration of the river mouth. A single major disruptive event located at 550 cm core depth and heralding the development of anoxia in the harbor marks the end of the dynamic regime that otherwise controlled the harbor water throughout the Roman Empire period. This remarkable

event may correspond to a major disruption of the aqueduct system or to a brutal avulsion of 33 34 the Cayster River bed. It clearly represents a major disturbance in the history of life at Ephesus. It is poorly dated, but probably occurred during the reign of Augustus or shortly after. Lead 35 isotope and trace metal evidence suggest that in the four bottom units pollution was subdued 36 with respect to other Pb metal inputs, presumably those from aqueducts and natural karstic 37 springs. Near the top of the core, which coincides with harbor abandonment and the more 38 39 recent period, anthropogenic Pb contamination is clearly visible in both Pb abundances and isotopic compositions. 40

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42 Keywords: Harbor geoarcheology, geochemistry, Pb isotopes, Roman age, paleo-pollution,
43 Ephesus, Küçük Menderes

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45 **1. Introduction**

Lead isotope studies have opened up a new, though somewhat controversial, perspective on the 46 47 development of the manufacturing status of ancient cultures over the past several millennia (Hong et al., 1994). Isolated artifacts alone do not suffice to assess the broad and long-lasting 48 aspects of antique trade routes. Lead isotopes constitute a complementary tool in that they play 49 50 a critical role wherever their compositions can be ascribed to anthropogenic influence in the form of lead and heavy metal pollution of sediments accumulated in harbors, which are highly 51 efficient traps for clays and suspensions. Anthropogenic impact using Pb isotopes as a tracer 52 has so far been documented for the ancient harbors of Alexandria (Véron et al., 2006; 2013; 53 Stanley et al., 2007), Sidon (Le Roux et al., 2003), Marseilles (Le Roux et al., 2005), and Rome 54 (Delile et al., 2014a). 55

56 Applying similar methods to the Roman harbor of Ephesus is appealing because of the 57 status of the Ephesus city port during Roman times as an exceptionally influential commercial

and religious center of the ancient Mediterranean world. Ephesus was a major town of Asia 58 Minor and has a long history that began in the 10th century BC.. Its position at short distances 59 from both the Dardanelles and the populated city states of southern Greece gave Ephesus a 60 strategic role in all the wars affecting Asia Minor and the Aegean Sea since the Persian wars of 61 the classical period. Its importance remained prominent during Hellenistic and Roman times 62 and during the entire history of the Byzantine Empire, and only declined as a result of the 63 64 Turkish conquest. Because sediments gradually filled in the inlet of the Cayster River (Küçük Menderes), the harbor of Ephesus repeatedly moved down river over the centuries (Kraft et al., 65 2000; 2011). 66

Here we use samples from a 12 m long sediment core taken in the Roman port of Ephesus to investigate the paleo-environmental and hydraulic evolution of the harbor using grain size distribution analysis, ¹⁴C ages, major and trace element geochemistry, and Pb isotope compositions. We focus in particular on the relative abundances of Pb and other chalcophile elements in the harbor sediments and discuss the respective status of the anthropogenic and natural metal fluxes and their origins as deduced from the Pb isotope record.

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74 2. Historical background

Literature on the history of Ephesus is abundant because of the wealth of ruins left by its different inhabiting cultures and its role in the history of this part of the world first as a major religious center dedicated to Artemis and later as one of the leading churches of the Mediterranean world. For a detailed historical context of the present work, the reader is referred to the well-documented textbook by Foss (1979) and to Scherrer (1995). Here we provide only a brief overview.

B1 Different sites were inhabited in the immediate vicinity of classical Ephesus since the
R2 Neolithic culture and during the Bronze Age. The historical city (close to the Artemision) was

founded in the 10th century BC by Ionians and became part of the Ionian League. The classic 83 84 site (at the base of the western side of the Panayırdağ) was occupied around 300 BC under Lysimachus, one of Alexander's generals, but quickly passed under Seleucid and then 85 Ptolemaic rules. After the Battle of Magnesia in 190 BC, Ephesus came under the domination 86 of Pergamon, and finally became part of the Roman Republic in 133 BC. After the Mithridatic 87 wars (ending in 63 BC), Augustus made Ephesus the capital of Asia Minor. At that stage, the 88 surface area of the city, enclosed by the walls of Lysimachus, is thought to have extended over 89 more than 2 km^2 and its population to have reached 50,000 inhabitants. 90

The city and its temple were destroyed by the Goths in 262 AD. But Ephesus was rebuilt 91 and enlarged by Constantine and soon recaptured most of the importance it had held since 92 Hellenistic times. A burst of seismic activity between 358 and 365 AD repeatedly destroyed 93 major cities around the Aegean (Guidoboni, 1994), including Ephesus. In the 7th century AD, 94 95 several additional disasters struck Ephesus, notably the major earthquake of 614 AD, as well as the repeated sacks by Arab, Frankish, and Turkish raiders. Western Turkey is well known for 96 being subjected to frequent earthquakes of large magnitude (e.g., Vannucci et al., 2004). 97 Although some dates are not well established, particularly severe earthquakes persistently 98 ravaged the city in AD 17, 23, 47, 178, 194, 262, 275, 337, 358 to 365, and 614 (Guidoboni, 99 1994; Foss, 1979). In AD 1304, what was by then left of Ephesus fell into the hands of the 100 101 Turks, and its population was either deported or massacred. These adverse troubles combined with the final stages of insilting of the harbor basin, which had incessantly plagued harbor 102 activity since its early Hellenistic days (Strabo, XIV.1.24), precipitated the demise of the harbor 103 and the city it served. 104

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106 **3. The study area**

107 Ephesus' harbor lies on the Aegean coast of Turkey at the western extremity of the Küçük Menderes graben (KMG) (Fig. 1). The KMG corresponds to the catchment area of the Küçük 108 109 Menderes (Cayster) river, which is divided into five sub-basins delimited by pre-Miocene geology (Rojay et al., 2005). The surrounding hills are composed of crystalline marble or 110 partially dolomitic breccias of Mesozoic age (Vetters 1989; Cakmakoğlu, 2007). The hills over 111 112 which Ephesus aqueducts run also include Paleozoic crystalline rocks such as granites, gneisses, and micaschists. Water was brought to the city by up to seven aqueducts built between 113 Archaic times and the Roman Empire and repaired during different periods, notably after major 114 115 earthquakes. This point is particularly important since all the waters from the aqueducts terminated in the harbor where they were susceptible to mixing with Cayster river water, marine 116 water, and waste waters of public (baths, fountains) and domestic usage, as well as with water 117 from local workshops (Ortloff and Crouch, 2001). 118

119 The variation of relative sea level and the westward migration of the shoreline since 120 Antiquity have been studied by Brückner (2005) and Pavlopoulos et al. (2012). Each century 121 the shoreline progressed westward by, on average, ~350 m. Comparison of the apparent sea 122 level changes with the values predicted by the regional model of Lambeck and Purcell (2005) 123 indicates that subsidence of the coastline next to Ephesus since the classical period was of the 124 order of 3 to 7 meters.

Geoarcheological research has been carried out at the Ephesus site and in the delta of the Cayster river since the 1990s (Brückner, 1997; 2005; Kraft et al., 1999; 2000; 2001; 2011; Stock et al., 2013; 2014). Besides reconstruction of the successive paleo-environments and the coastline as it has existed since 6000–5000 BC (Fig. 1), this work also has shown that delta progradation led to multiple westward resettlements of the harbor. The ceaseless fight against silting to maintain the harbor of Ephesus as a functioning port during the Hellenistic period is first and foremost reflected in the displacement of the city to the western side of mount Pion
(Panayırdağ) by Lysimachus in ~290 BC (Scherrer, 1995).

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134 4. Analytical techniques

A sediment core about 12 m long (EPH 276) was drilled in the hexagonal Roman harbor basin 135 of Ephesus (Fig. 1). We sampled the core at high resolution by taking a total of 111 samples 136 (one sample every 10 cm). The samples were analyzed for grain size distributions (see Stock et 137 al., 2013, for details), major and trace element concentrations (Table S1; see Delile et al., 2014b, 138 for details), and Pb isotope compositions (Table S2; see here below and Delile et al., 2014a, for 139 140 details). Lead isotope compositions were obtained not on the bulk sediment, but, in order to isolate potential anthropogenic components, on HBr leachates. The leaching procedure 141 consisted in first treating the samples with chloroform to remove most of the organic fraction, 142 143 then, after rinsing the residues with clean water, leaching them with dilute HBr including ultrasonicating and heating steps. As shown for Portus (Delile et al., 2014a), this technique 144 145 enhances the contrast between Pb held in surface contamination-prone coatings and detrital 146 silicates. Carbonates also dissolve during the leaching process, but Pb contents of detrital carbonates are naturally low. As for carbonates precipitated within the harbor, they are of 147 biogenic origin (cf. discussion of series D and E below) likely meaning that isotope information 148 obtained on carbonate-rich samples is consistent with that derived from the leachates of the rest 149 of the sample series. The HBr leach fraction was recovered for Pb separation by anion-exchange 150 chromatography using HBr as eluent of the sample matrix and HCl as eluent of the Pb. Lead 151 was also separated from the residues of 16 of the 111 EPH 276 samples. The amounts of Pb 152 extracted from all samples were large (>1 μ g) and orders of magnitude above the total 153 154 procedural blank of ~20 pg. The purified Pb was analyzed for its isotopic composition by multiple-collector inductively coupled plasma mass spectrometry (Nu Plasma 500 HR) at ENS 155

Lyon using Tl for instrumental mass bias correction and bracketing the samples with the NIST981 standard for which the values of Eisele et al. (2003) were used.

Six samples were AMS-¹⁴C-dated (Table S3), complementing the chronostratigraphy of 158 neighboring cores analyzed by Stock et al. (submitted). The Carbon-14 ages were obtained on 159 fragments of wood, vegetal matter, seeds, and pollen, and are listed in Table S3 and shown in 160 Figs. 2 and 3. Errors on raw radiocarbon ages BP are reported at the 95 percent confidence level 161 (two sigma). The measured ${}^{14}C$ (BP) ages were converted into BC – AD dates relative to the 162 continental and marine curves of Reimer et al. (2009) using the Clam software (Blaauw, 2010). 163 Interpretation of the analytical results rests on different methods of data processing. We 164 165 applied Principal Component Analysis (PCA) and Factor Analysis to major and trace element concentration data, as well as loss-on-ignition (L.O.I.) (Fig. 2). In the very large data sets typical 166 of those that modern geochemistry can now produce, observations are often correlated. A 167 168 common case is that of the dilution of elements in sediments by detrital quartz. Such effects render the reading of the underlying causes of geochemical variation and their number difficult. 169 170 PCA consists in rotating the data in their multidimensional space to convert them into uncorrelated variables known as principal components. Uncorrelated does not equate with 171 independent, however, implying that small changes in rotation may affect all the principal 172 components. PCA generally demonstrates that the variability of the observations can be 173 accounted for by a very small (2-4) subset of variables that carry the bulk of the total variance. 174 Principal components can be calculated from the covariance matrix or from the correlation 175 matrix. Factor Analysis is a related technique that searches for the minimum variance for an 176 arbitrary number of uncorrelated variables. It usually starts with PCA and implements different 177 modes of rotation and weighing. 178

179 In addition to the PCA and Factor Analysis we also converted the Pb isotope 180 compositions into their corresponding geochemically informed parameters, which are the

model age $T_{\rm mod}$ and the ²³⁸U/²⁰⁴Pb (μ) and Th/U (κ) ratios (Table S2) using the equations given 181 by Albarède et al. (2012), who also justified the advantages of this representation over those 182 183 based on raw Pb isotope ratios. In short, T_{mod} is a proxy for the tectonic age of crystalline rocks and their associated ore deposits (or depositional age for sediments), while μ is the $^{238}\text{U}/^{204}\text{Pb}$ 184 and κ the Th/U ratio of the province in which these rocks formed. T_{mod} closely maps the 185 distribution of the Alpine, Hercynian, and early Paleozoic provinces of Europe, while 186 μ delineates collision belts, and κ is a geochemical parameter with a remarkable regional 187 consistency related to uplift and erosion. Maps of these parameters can be used to divide Europe 188 into coherent regions (Delile et al., 2014a), which justify the use of T_{mod} , μ , and κ to determine 189 provenance of archeological artifacts. T_{mod} , μ , and κ in turn provide a rapid characterization of 190 the geological environment in which ores formed. A Matlab code is given in Appendix A and 191 an Excel spreadsheet in which to calculate these parameters will be provided upon request. As 192 193 mentioned above, T_{mod} represents the tectonic age of the geological province to which a given sample belongs, while μ is best perceived as an indicator of whether this province is a collision 194 range or a tectonically stable area. The variable κ distinguishes upper crust with low κ values 195 from middle and lower crust with higher κ values (Albarède et al., 2012). The precision and 196 accuracy of $T_{\rm mod}$ is typically of a few tens of Ma, but, occasionally, the $T_{\rm mod}$ - μ - κ model fails 197 when the underlying closed-system assumption breaks down due to U addition by recent 198 weathering or hydrothermal activity. 199

- 200
- 201 5. Sedimentary units and the age-depth model

The core has been divided into five different units labeled A, B, C, D, and E on the basis of the sedimentological and geochemical traits described in Fig. 3A; they span the entire period of activity of the Hellenistic, Roman, and Byzantine harbor (Table 1).

Unit A (1200-1080 cm) exhibits alternating brown and grey varves composed of 205 206 massive clayey silts with the presence of several beige to ocher fine layers. The C/M plot (Fig. 3B) indicates that the depositional processes are represented by mixed decantation and graded 207 suspension. Units B and C (1080-515 cm) are characterized by grey to greenish massive sandy 208 silts with the presence of several beige to ocher fine layers enriched in sand. This deposit was 209 deposited as a graded suspension with embedded fine layers derived from mixed processes of 210 graded and uniform suspension. Unit D (515-290 cm) consists of dark to greyish silts with 211 212 variable clay (bottom) and sand (top) enrichments. From bottom to top, deposit processes evolve from mixed uniform and graded suspension to a blend with graded and uniform 213 suspensions. Unit E (< 290 cm) is composed of beige sandy silts with phragmite vegetal 214 remains. The depositional processes point mostly to mixed graded and uniform suspensions. 215

The age-depth model is based on six ¹⁴C ages (Fig. 4, Table S3). The four oldest ¹⁴C 216 217 ages fall within a narrow time interval and are statistically indistinguishable. An approximate seven meters of sediment were deposited in a few tens of years during the reign of Augustus or 218 219 shortly after. Such an extraordinarily fast sedimentation rate is consistent with a periodogram 220 analysis (e.g., Albarède, 1995) of the magnetic susceptibility record. The periodogram, which is the equivalent of a Fourier transform for unequally spaced data, identifies prominent periodic 221 fluctuations in the targeted property, here the magnetic susceptibility. After removal of long-222 223 term variations (de-trending) by fitting a fourth-degree polynomial, the shortest values with significance level P >0.95 occur at 20 and 24 cm (Fig. 5). Longer wavelength peaks probably 224 reflect climatic effects or are artifacts of de-trending. Assuming a seasonal cause for the 225 observed susceptibility fluctuations therefore indicates a sedimentation rate of ~20 cm per year, 226 equivalent, over the 7 meters of sediment with the oldest ${}^{14}C$ ages, to ~35 years of sedimentary 227 history. In contrast, the average sedimentation rate between the top three ¹⁴C samples (early and 228 late Byzantine) is only ~ 0.2 cm a⁻¹. 229

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231 6. Results and discussion

232 6.1. Harbor hydraulics

In order to understand the hydraulic dynamics of the harbor, we first need to estimate its water 233 capacity. The approximate dimensions taken from aerial photographs lead to a volume of (500 234 $\times 400 \times 5$) m³ = 1.0 $\times 10^6$ m³ (see also Kirbihler, 2013; Stock et al., submitted). We assume that 235 236 most of the sedimentary layers were deposited during short seasonal flood events of the Cayster 237 River, whereas water running into the harbor came from different potential sources: seawater, runoff, karstic springs, and aqueducts. Seawater and water from springs and aqueducts must 238 239 have been largely clear of sedimentary particles. Karstic springs are common in the area around the modern Lake Kocagöz (Somay et al., 2008; Somay and Gemici, 2009), which today exists 240 on the site of the ancient harbor basin. A seawater component is present in the water from all 241 242 the lakes, including Lake Kocagöz (Somay et al., 2008; Somay and Gemici, 2009). Such a component attests to a contribution from spring waters contaminated by marine intrusions into 243 244 the karst. As for other water inputs, Kraft et al. (2007) pointed out that all the city sewage was diverted into the Great Harbor. 245

The up to seven aqueducts built during the existence of the Ephesus port carried 246 substantial volumes of water into the harbor. Wiplinger (2013) quotes an estimate of 0.6 m³ s⁻¹ 247 for the Derğirmendere aqueduct alone. Using the model by Ortloff and Crouch (2001), we 248 surmise that the total water distribution to the city from the fully-functional aqueducts at the 249 peak of city prosperity may have been over $\sim 2 \text{ m}^3 \text{ s}^{-1}$. This number is substantial with respect 250 to the mean discharge of ~11.45 m³ s⁻¹ inferred for the river, not including flood events 251 (Vliegenthart et al., 2007), indicating that if other inputs such as karstic springs and runoff are 252 disregarded, water in the harbor was replaced by the aqueducts in merely six days. This estimate 253

is of course an average estimate and during seasonal droughts the ingress of seawater attestedto by the presence of brackish fauna also contributed to the harbor's overall water budget.

Water output is difficult to constrain independently. The harbor canal, whose construction 256 257 may have started as early as during the first century AD when the shoreline swept past the harbor, was narrow at the harbor entrance (Kraft et al., 2007). Assuming a cross-section at the 258 narrowest point of $\sim 50 \text{ m}^2$ would imply that aqueduct-delivered water was leaving the canal at 259 a rate of 145 m per hour, probably fast enough to limit water ingress from the sea under fair 260 weather conditions. This velocity must have been reduced by the effect of draught and 261 evaporation during the dry (hot) season, and increased by local springs during the wet (cold) 262 263 season. The presence of brackish water ostracods and occasional occurrence of marine foraminifera (Stock et al., submitted) demonstrate that the flow could occasionally be reversed, 264 presumably as a result of a low water table and reduced precipitation during the dry season. 265 266 Over time, the harbor was nevertheless affected by westward delta progradation and proximity to the mouth of the Cayster river (Fig. 1): by the end of the 2nd century BC, the delta had 267 advanced as far as the Great Harbor (Kraft et al., 2007) and the canal had to be extended, thus 268 269 limiting ingress of seawater into the harbor basin even further.

To summarize harbor hydrodynamics, the 'normal' situation is that of a basin steadily 270 filled by polluted urban water initially brought to the harbor by aqueducts and local springs and 271 quickly evacuated through a canal with little ingress from the sea. As long as the coastline is 272 not too distant, some seawater may be admitted during the dry season, while floods of, in the 273 present case, the Cayster River dominated the water balance during spells of heavy rain. Silting 274 in of the harbor would have been caused only by floods, which today are known to carry up to 275 100–150 m³ s⁻¹ of water (Vliegenthart et al., 2007). The Romans went to great length to protect 276 the harbor from river floods. Kraft et al. (2000; 2011) mention that, in the early 2nd century AD, 277

Hadrian sought to divert the Cayster River with an 18 m high dam and also made multipleattempts to dredge the harbor.

- 280
- 281 6.2. Environmental conditions in the harbor basin

In order to assess the environmental conditions that prevailed during sedimentation, we plotted the concentrations of first-row transition elements (Ti-Zn) and other metallic elements (Ga, Pb, Mo, Bi, Cd, Ag, As, and Sb) normalized to the upper-crust concentrations of Rudnick and Gao (2003) (Fig. S1). Factor analysis of major and trace element abundances leads to the identification of three major components.

- *The first factor* opposes elements indicative of the detrital load of the river (Al, Ti, Mg, etc.) to those distinctive of carbonate minerals (Ca, Sr) and L.O.I.
- 2. The second factor singles out chalcophile elements that, as attested to by the presence 289 290 of sulfur in this group, precipitate as sulfide under anoxic conditions (Pb, Ag, Cu, Ni, Mo), or are particularly sensitive to redox conditions (U, Cr). When the elements 291 embedded in this factor are normalized to Al (Fig. 6), as a means of accounting for the 292 variable abundance of the detrital component, and plotted against depth in the core, a 293 sharp increase is observed at 520 cm depth. The significance of this factor deserves 294 some discussion because Pb, Ag, and Cu may also be seen as representing an 295 anthropogenic component. Figures 6 and S1 further show the striking consistency of 296 these metals among themselves and with respect to sulfur. Such regular behavior is not 297 supportive of random contamination by a particular metal, such as Pb. The Mo-Pb 298 correlation is very strong (r=0.90 excluding the top five samples likely contaminated by 299 gasoline Pb) as is the Ag-Pb correlation (r=0.95). This factor therefore reflects more on 300 changing redox conditions in the harbor than on anthropogenic pollution. 301

302 3. *The third factor* is dominated by La, Ce and, to a lesser extent, Se. Most other loadings
303 are very small, except possibly P. The weak negative correlation between excess La and
304 Ce on the one hand and P deficit on the other hand suggests the presence of non305 phosphatic rare-earth minerals, such as allanite, notably in the coarse silts between 515
306 an 650 cm.

307 The accumulation of so much sediment in a matter of decades requires an explanation, especially since the thickness of the newly deposited layers exceeds the water depth usually 308 assumed for the harbor (4-6 m), even next to the mole (Stock et al., submitted). One factor 309 clearly is the westward progradation of the shoreline past the harbor at about the time of fast 310 sedimentation. F. Stock (personal communication) obtained a ¹⁴C age of 44 BC – AD 52 for 311 312 the silting in of the harbor canal consistent with the present finding. The sharp geochemical discontinuities at 650 and 550 m are flagged by strong peaks of magnetic susceptibility (Fig. 313 3A). The efficiency of sediment confinement by the harbor prior to the three lowermost units 314 is staggering, while the sudden drop in sedimentation rate and the short but intense episodes of 315 high magnetic susceptibility require the intervention of a brutal event. A probable cause for this 316 discontinuity is an abrupt jump of the Cayster River channel triggered by the abandonment of 317 a meander (avulsions) or by exceptional floods (Brown, 1997). Co-seismic vertical movements 318 319 (Pavlopoulos et al., 2012) associated with the major AD 17, 23, and 47 earthquakes may also have played a role. 320

The lowermost *unit A* (1200–1080 cm) was deposited during the Roman Republic. It is consistently dominated by silt (F1>0) with anoxic influence (F2<0). The anoxic conditions of the basin bottom as attested to by abundant S, Mo, and U (Fig. 2), small excesses of Mo and Ag (Fig. S1), and persistence of seasonal varves, indicate that the terrigenous flux into the early Roman harbor of Ephesus during the 1st century BC was not noticeably oxidized whether water input was freshwater or seawater. Grain size distribution (Fig. 3) reflects an environment where decantation is important (Bravard and Peiry, 1999; Bravard et al., 2014). Input of oxygenated
freshwater into the harbor, regardless of its source, therefore was limited and whatever water
was added by the aqueduct system must have been dominated by sewage.

Unit B (1080–650 cm) continues to show the prevalence of the detrital flux (F1 >0), but 330 now with evidence of oxygenation (F2 \geq 0). The transition-element pattern typically is crustal 331 in origin and no visible anomaly of Mo and Ag is observed (Fig. S1). Grain-size analysis 332 indicates graded grain size distributions by turbulent waters, reflecting that, even at times of 333 flood, water was being constantly evacuated from the harbor. Ephesus counted up to seven 334 aqueducts implying that the early Roman harbor was saved from silting in as much by water 335 336 from its many aqueducts continuously flushing the basin as by the Roman engineers. As shown by the return of some decantation events (Fig. 3), the aqueducts made silting depend on a fully 337 functioning water distribution system. In this respect, the Menderes area is seismically active 338 339 (Vannucci et al., 2004) and major earthquakes were particularly disruptive to the long and complex Ephesus aqueduct network (Passchier et al., 2013). Reduction of the water input by 340 341 the seismic destruction of aqueducts translates into reduced water egress from the harbor basin and hence enhanced efficiency of its role as a sediment trap. Silting in of the harbor in the 342 aftermath of major earthquakes therefore became collateral damage to the rest of the disasters 343 caused by the seismic activity. 344

Transition to *unit C* (Fig. 2) (650–550 cm) is heralded by a peak of magnetic susceptibility (Fig. 3A). Highly negative values of factor 3, i.e., higher Se, La, and Ce contents, reflect lesser dilution of minor elements by quartz and carbonate. The variation patterns of transition elements and other metals are very similar to those of unit B. As already observed for rivers (Yang et al., 2002), a strong correlation exists between grain size and lanthanide concentrations (Zhang et al., 1998; Yang et al., 2002). This geochemical change is consistent with a sand fraction in unit C smaller than that in unit B. Unit C shows some transient geochemical features (Fig. 7), true harbingers of the major changes that would profoundly affect unit D, notably anincrease in sulfur and heavy metal contents

The transition (550-515 cm) between unit C and unit D (515-300 cm) also is announced 354 by a strong peak of magnetic susceptibility (Fig. 3A), corresponding to a strong compositional 355 shift with a surge of the biogenic component (F1<0) due to degraded ventilation of bottom 356 waters by eutrophication (F2<0). Sulfide reduction and precipitation is attested to by a sudden 357 358 two-order-of-magnitude increase in the S/Al ratio (Fig. 6). The surge in sulfur, Zn, Ni, and Co 359 conspicuously follows the surge of Ca, Pb, Ag, Cu, Cd, Mo, and Cr by some 30 cm in the core. This delay, which may have been as short as a few years and possibly was only one or two 360 years, is visible in the plot of Fig. 7 as a pronounced negative excursion of ratios such as Ca/S, 361 Mo/S, and Pb/S between 520 and 550 cm depth. These characteristics together with high Sr 362 abundance and the presence of fine calcareous layers (Kylander et al., 2011; Martín-Puertas et 363 364 al., 2011) show that sulfide precipitation predated the development of eutrophic conditions manifested by the rise in Ca and was due to the sudden isolation of the harbor from ventilated 365 waters. The trend of decreasing ratios of chalcophile elements relative to sulfur, which was 366 perceptible already in the early harbor, markedly changes at the C-D boundary, and the rate at 367 which these ratios change significantly increases as well. Again, the correlation between Cr, 368 Cu, Pb, Mo, and Ag (Figs. 6, 7, S1) excesses are not in favor of selective pollution by 369 370 metallurgical or any other industrial activities. The destruction of the aqueducts by the major earthquakes ravaging the city, such as the AD 17, 23, and 47 events (see a discussion in 371 Guidoboni, 1994), and the AD 64 AD cleaning credited to Barea Soranus by Tacitus (XVI,23) 372 373 may have combined with the increasing silting of the harbor entrance upon westward progradation of the delta (Fig. 1) (Brückner, 2005) to modify the hydraulic regime of the harbor. 374 The top of unit D records a short-lived return of better oxygenated conditions which, with the 375

caution due the age-depth model, may correspond to the revival of the harbor by Justinian (early
and mid- 6th century; Foss, 1979; Scherrer, 1995).

The age-depth model (Fig. 4) places the transition between units D and E (~300 cm) in 378 the 9th century. Carbonate precipitation dominates unit E as it did the lower part of unit D (F1<0) 379 indicating a negative water balance (Fig. 2B; Martín-Puertas et al., 2011; Delile et al., 2014b). 380 Excesses of Cr, Cu, Mo, and Ag are still well correlated (Fig. 2A) and confirm the persistence 381 of a sulfur-rich, oxygen-deficient eutrophic regime, but, as shown by the positive F2 values, 382 with oxygen deficiency being less pronounced than in the underlying unit D. The water deficit 383 caused conditions to evolve towards a peatland environment consistent with the considerable 384 385 extension of the Cayster delta at this time (Fig. 1). The modern estuary of the Küçük Menderes is wetland dotted with alkaline lakes recharged from precipitation and local karstic springs. In 386 late Byzantine times, the harbor may have been functional, but appears to have been 387 388 increasingly cut off from the sea and the river (Kraft et al., 2011) (Fig. 1). Some of the shallow core samples show excess Pb of probable but uncertain anthropic origin (Fig. 8). 389

To sum up on environmental conditions, core EPH-276 holds the record of anoxic conditions prevailing at times in the harbor, likely compromising the control of harbor hydrodynamics by human activities. Lead isotopes are expected to shed light on the magnitude of anthropogenic contamination at the time of sediment deposition and this is what the next section will be addressing.

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396 6.3. Interpretation of Pb abundance and isotopic signals

Here we focus on the Pb isotope compositions of the leached fractions only because this is where chances of observing anthropogenic input are maximum. Figure 8 shows the Pb/Al ratio together with the Pb isotope data in the form of three geochemical parameters, T_{mod} (Ma = million years), μ , and κ . Based on these parameters and the enrichment factor of Pb as

represented by Pb/Al and closely tracking F2, the chronostratigraphic evolution of these four 401 402 curves shows remarkable discontinuities (Fig. 2). In agreement with what was discussed above for other metals, the transition between units A and B stands out clearly by a marked drop in 403 Pb/Al. A subtle increase in μ and κ , while T_{mod} remains young (~80 Ma or Upper Cretaceous), 404 is evidence of change in the sources of Pb. The next discontinuity takes place between units C 405 and D. The increase in Pb/Al and Ca/Al is associated with older T_{mod} (~120 Ma or Early 406 Cretaceous) and lower values of μ and κ . The Pb/Al ratio decreases steadily throughout unit D 407 regardless of the changes in the major Ca/Al dip at 3.8 m depth that we assigned above to the 408 6^{th} century. From unit D to E, most T_{mod} values exceed 240 Ma and the κ values decrease below 409 the level of previous values. The samples at the top of the core seem to be largely influenced 410 by a modern anthropogenic component. 411

The hydraulics of the harbor, notably its volume and output, may affect harbor oxygenation and thereby the metal contents of sediments. In contrast, changing Pb isotope compositions require changes in the relative contribution of all the sources of this metal. Lead isotopes reveal the nature and relative strengths of the following potential sources:

Local natural sources, which are multiple. Lead from the Cayster River comes during
short-lived seasonal flood events. Seawater should be extremely poor in Pb, while
brackish water from the estuary should be Pb-depleted by iron flocculation in the
mixing zone. A contribution from the runoff and from karstic springs that discharge
from marble-schists and marble-alluvium contacts (Somay et al 2008; Somay and
Gemici 2009) should also be considered.

Lead from the main water distribution system. Such a component may come from the
underground of the springs. It can also be acquired during transit from the aqueduct
masonry, which includes mortar produced from local limestones. The laminated
deposits observed in some aqueducts (Passchier et al., 2011; 2013) indicate hard water

with excess alkalinity, which does not favor the idea that Pb was leached out of these
conduits. The seemingly high concentrations of chalcophile elements in the sediments,
first and foremost Pb but also Co and Mo, do not entail a pollution signal.

3. The network of aqueducts, which is unlikely to be a major Pb contributor. The 429 secondary water distribution system at Ephesus is dominated by terra cotta pipes and 430 only rarely involves small-diameter lead pipes or fistulae (Ortloff and Crouch, 2001). 431 Anthropogenic Pb from local workshops or ballast dropped by merchant ships. 4. 432 Zabehlicky (1995) writes that a lead anchor 14.2 cm long was found during 433 excavations, as well as lead interpreted as ballast, which as much as hinting at a 434 435 potential source of pollution, signals that the dissolution of Pb artifacts is an exceedingly slow process. The presence of an arsenal on the harbor site at the time of 436 Augustus was noted by Strabo (XIV.1.24). There is no doubt that a city with the 437 population of Ephesus at its best periods would have to rely on local metallurgy. In 438 sediments, however, a geochemical signal of pollution is difficult to detect, and to 439 which extent Pb artifacts attest to wholesale contamination of harbor sediments is not 440 clear. The stability of Pb isotope compositions over long periods of time (decades, 441 even centuries) does not bring to mind metal supply in troubled times. Even more 442 443 conclusive is the observation that, except for unit E, Cr, Cu, Mo, Ag, and Cu coherently track Pb and Ca: although these elements are sensitive to redox conditions, they were 444 not involved in Pb metallurgy. The Pb/Ag ratio remains remarkably stable, while the 445 record of Ni and Co, for which extractive metallurgy was unknown at the time and 446 which additionally are not found in the same ores as Pb, also follow the Pb record with 447 depth. The lesser impact of anthropogenic Pb pollution at Ephesus relative to Portus 448 (Delile et al., 2014a) directly reflects that the urban water distribution systems used 449

450

451

different materials, *terra cotta* for the former (Ortloff and Crouch, 2001) and lead fistulæ for the latter.

The Cretaceous model ages of 80 Ma and 120 Ma observed in the lower part of the core are 452 453 consistent with those of the carbonate hills surrounding Ephesus and may simply register Pb from the water distribution system (natural springs or conduits). This interpretation is supported 454 455 by the similar Pb isotope compositions of leachate-residue pairs (Table S2). The residues of the samples with T_{mod} values >200 Ma have not been analyzed, but these ages are consistent with 456 the age of the Paleozoic and Triassic crystalline basement of the Menderes Massif (Vetters, 457 1989; Bozkurt, 2007; Çakmakoğlu, 2007; Gürer et al., 2009) (see maps in the supplementary 458 material of Delile et al., 2014a). 459

The appearance of old $T_{\rm mod}$ values shortly after the beginning of unit E, which have not 460 been observed at earlier times, coincides with the onset of carbonate sedimentation (up to 45 461 wt.% CaO). Runoff and karstic springs more or less contaminated by seawater (Somay et al 462 2008; Somay and Gemici 2009) are therefore left as the main steady sources of water in the 463 ancient port, which since has become the modern Lake Kocagöz. Two competing 464 interpretations are left: (1) while ancient aqueducts were bringing in Cretaceous Pb, runoff and 465 karstic springs now bring in Paleozoic Pb; or (2) old T_{mod} values reflect some anthropic 466 influence of poorly constrained origin. The younger samples with T_{mod} values >200 Ma may 467 represent sources in the Menderes region, but Pb from Thrace, or even from western Europe, 468 469 cannot be excluded.

Historical evidence in favor of Ephesus hosting significant industrial activity in the 11th
and 12th centuries (Foss, 1979, pp. 120-123), however, is faint. Foss (1979, p. 113) further
argues that from the 8th century onwards the harbor district was literally abandoned. The center
of town moved to the hill of Ayasuluk, while new ports, such as Scalanova (ancient Phygela)
built on the site of modern Kuşadası, gradually took over the silted in harbor of Ephesus.

475 Nevertheless, from the middle of the 9th to the 10th century, the victories of the 476 Byzantines against the Arabs in Asia Minor enabled Ephesus to regain a preeminent position 477 in the Empire (Foss, 1979). A phase of economic development would be consistent with the 478 influx of Hercynian Pb into the most recent harbor deposits. The 10th century medieval 479 economic revolution in Europe favored the development of trade between Europe and the 480 Orient. West or north European sources of Pb cannot be excluded for that period.

481

482 **7. Concluding remarks**

Major changes in the lithology, grain-size distribution, major and trace element chemistry, and Pb isotope compositions of the harbor sediments at Ephesus reflect the history of the water distribution system of this port, notably in response to the increasing and declining needs of a population inhabiting a city that at several points in history was one of the largest of the Eastern Mediterranean. Throughout its history, the Ephesus port was affected by major disruptive events in the form of earthquakes and invasions, both of which were particularly effective at destroying aqueducts.

Progressive silting in of the harbor responded to the westward migration of the coastline 490 and to human maintenance aimed at keeping the harbor functional. A single major disruptive 491 event located at 550 cm core depth and heralding a two-order-of-magnitude drop in 492 sedimentation rate and the development of anoxia in the harbor is clearly visible in the major 493 and trace element record. Although this event may have unidentified military or seismic causes, 494 we favor a durable displacement of the river course, which starved the harbor from further silt 495 496 input. Overall, despite the presence of metallic artifacts in the harbor, the record of metal concentrations, in particular the Pb isotope record, suggests that pollution of the harbor was 497 498 subdued relative to other inputs, notably those of aqueducts, except near the time of harbor abandonment (unit E). 499

Dating and identifying the seemingly key event located in the present sediment core at 501 550 cm depth, as well as in other cores from the same basin, is a new and major challenge. This 502 event conspicuously marks the end of the dynamic regime controlling the harbor water during 503 all of the Roman Empire and clearly represents a major disturbance in the history of life in 504 Ephesus.

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- 506

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517 Appendix A: Matlab code for T_{mod} , μ , and κ calculations

The input data should be provided as an Excel file input.xlsx made of three columns,holding ²⁰⁶Pb/²⁰⁴Pb,
 ²⁰⁷Pb/²⁰⁴Pb, and ²⁰⁸Pb/²⁰⁴Pb.

- 520
- 521 % Matlab code for model age calculations (Francis Albarede)
- 522 %
- 523 A=xlsread('input.xlsx',1);
- 524 m=size(A,1); % find m the number of samples
- 525 % decay constants
- **526** 18=0.155125;15=0.98485;12=0.049475;
- 527 % common Pb parameters from Albarede and Juteau (1984)
- 528 xstar=18.7500;ystar=15.63;zstar=38.83; kappastar=3.90;mustar=9.66;
- 529 % Initialize
- 530 FF=zeros(m,1);mu=zeros(m,1);dmu=zeros(m,1);kappa=zeros(m,1);dkappa=zeros(m,1);Tmod=zeros(m,1);Tinit=zeros(m,1);dmu=zeros(m,1);kappa=zeros(m,1);dmu=zer
- 531 1;

532	T0=3.8; % Beginning stage 2 (Ga), see Stacey and Kramers (1975)
533	options=optimset('Display','off');
534	for i=1:m % loop through all samples
535	x=A(i,1); y=A(i,2); z=A(i,3);
536	myf = @(T)TmPb(T,T0,x,y);
537	[Tmod(i),FF(i),exitflag(i,1)]=fsolve(myf,Tinit,options); % use Matlab fsolve with function TmPb
538	T1=Tmod(i);
539	dmu(i)=(x-xstar+mustar*(exp(18*T1)-1))/(exp(18*T0)-exp(18*T1));
540	mu(i)=mustar+dmu(i);
541	dkappa(i)=(z-zstar+mustar*kappastar*(exp(l2*T1)-1)-kappastar*dmu(i)*(exp(l2*T0)-
542	exp(l2*T1)))/(exp(l2*T0)-exp(l2*T1))/mu(i);
543	kappa(i)=kappastar+dkappa(i);
544	end
545	B=[A,1000*Tmod,mu,kappa,FF,exitflag]
546	% B holds columnwise the original ratios (A), the model age (in Ma), mu, kappa, the
547	% exit value of the function to solve (should be less than 1e-9) and an
548	% exit parameter (1 expected)
549	
550	function $F = TmPb(T,T0,x,y)$
551	mustar=9.66;
552	xstar=18.7500;ystar=15.63; %AJ
553	18=0.155125;15=0.98485;
554	p0=(exp(15*T0)-exp(15*T))/(exp(18*T0)-exp(18*T))/137.8;
555	p1=(exp(15*T)-1)/(exp(18*T)-1)/137.8;
556	%
557	if x~=xstar % Eq. 12 in Albarede et al. (2012)
558	F=(y-ystar)/(x-xstar)-p0-mustar*(exp(18*T)-1)*(p0-p1)/(x-xstar);
559	else
560	$F=(y-ystar)-mustar^*(exp(18*T)-1)^*(p0-p1);$
561	end
562	end
563	

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- 715

716 Figure captions (see below for figs)

Fig. 1. Map of the Küçük Menderes graben located on the Aegean coast of Turkey (inset) with
successive positions of the shorelines and site of core EPH 276 (from Brückner, 2005, modified
by Stock et al., 2013 and this study).

Fig. 2. Factor analysis of major and trace element concentrations. Based on Principal 720 Component Analysis, the number of factors is limited to three. (A) Component loadings. The 721 first factor F1 shows the trade-off between an (Al, Ti)-rich detrital component (F1>0) and a Ca-722 rich carbonate component (F1<0). The second factor F2 is dominated by metals (Ag, Pb, Cu, 723 Mo) and sulfur and shows the effect of anoxia. The third factor F3 is dominated by the light 724 725 rare-earth elements La and Ce and testifies to the presence of heavy minerals in sand. (B) Distribution of the different factors with depth in the column. The plots are compared with the 726 sedimentary units and the age-depth model. 727

- Fig. 3. (A) Stratigraphy and sedimentology of core EPH 276 showing grain size distribution and environmental facies. (B) Plot of the grain size 99 percentile (D 99) versus the median size (D 50) for the different samples analyzed. The different color groups correspond to different sedimentation regimes (see legend).
- Fig. 4. Age-depth model for core EPH 276 deduced from the six ¹⁴C dates with ranges calculated using the Clam software (Blaauw, 2010). The size of the data symbols reflects the confidence level.
- Fig. 5. Periodogram of magnetic susceptibility in core EPH 276 between 440 and 1100 cm. 735 Mean sampling interval is generally ~1 cm. Four extreme values corresponding to 736 737 discontinuities were removed from the data set. A fourth-order polynomial was then fitted to the data to remove the long-term trend. Peaks correspond to dominant periods, with confidence 738 levels P in percent in parentheses. We consider that the shortest periods with $P \ge 95\%$ 739 740 correspond to dominant annual varves, while longer periods correspond to climatic effects or to de-trending artifacts. The periodogram is interpreted as indicating an average sedimentation 741 rate of 20 cm per year. 742
- Fig. 6: Downcore variations of calcium and metal concentrations normalized to aluminum. White- and grey-shaded bands delineate the stratigraphic units A–E, with unit A being deepest and unit E shallowest. Note the discontinuities at ~1080 and ~550 cm depth, notably the increase in S and chalcophile elements at the latter.
- Fig. 7. Downcore variations of calcium and metal concentrations normalized to sulfur (see 747 748 caption of Fig. 4 for details). Two major discontinuities are again observed at ~1080 and ~550 cm depth, which attest to sudden changes in sediment oxygenation. The 1080 cm discontinuity 749 reflects the improved oxygenation of the harbor in the early 1st century AD. Most elements 750 show a negative excursion between 550 and 520 cm indicating that the rise in sulfur precedes 751 the rise in Ca and most other metals; exceptions are Zn and Co, which are in phase with S. 752 753 Although a slow trend towards anoxia can be seen in sediments below the 550 cm level, the 754 rate of ventilation is greatly reduced after this episode.
- Fig. 8. Downcore variations of Pb/Al_{norm}, T_{mod} , μ (²³⁸U/²⁰⁴Pb), and κ (Th/U) compared with variations in Ca/Al and Mo/Al. Pb/Al is normalized to the upper continental crust average of McLennan (2001). Major T_{mod} discontinuities are observed between units C and D, and at the base of unit E. A major discontinuity in both μ and κ is observed between units A and B. The

variability within unit E reflects an anthropogenic component of uncertain origin.

760

761 **Table captions**

Table 1. Depth range of the sedimentary units and probable age assignment.

763

764 Supplementary figure caption

Fig. S1. Abundances of the first-row transition elements (left) and of other metals (right) normalized to the upper crust average of Rudnick and Gao (2003). The different panels correspond to the five sedimentary units (A to E) as identified by sedimentology. Units B and C are smooth, which indicates the predominance of detrital input. Unit A and, to a much greater extent, units D and E show excesses of Mo, Pb, and Ag, which reflect the prevalence of anoxic conditions. Units D and E also show excesses of Cr and Cu. In unit E, the decoupling of Pb from the other elements attests to the addition of anthropogenic Pb (likely present-day gasoline).

772

773 Supplementary table captions

- Table S1. Major and trace element data for the core samples.
- Table S2. Lead isotope data for the core samples.
- Table S3. Radiocarbon results for core EPH 276.



Fig1











А















Fig.	8
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Unit	Depth (cm)	Probable Age					
A	1200-1070	Roman Republic					
В	1070-650	Early Roman Empire					
С	650-515	Late Roman Empire					
D	515-300	Early Byzantine (4th-8th century)					
Е	300-0	Late Byzantine and Turkish					

Table 1 : Depth range of the sedimentary units and probable age assignment.





Table S1. Major elements are in wt.% and trace elements are in ppm.

Lab code 129L	Depth (cm) 50	Unit Al ₂ O	CaO 13.49	Fe ₂ O ₃ 2.73	K ₂ O 0.92	MgO 2.19	MnO 0.041	Na ₂ O 0.82	P ₂ O ₅ 0.26	SO ₂ 1	TiO ₂ L	OI .97 3	Zr Li 33.67 19.	Be	V 61.04	Ga 7.87	Se 2.34	Rb 78.05	Sr 199.01	Y 10.46	Mo 3.70	Ba 151.85	La 14.34	Ce 28.74	Nd 12.13	Sm 2.53	Eu 0.56	Gd D	y Er	Yb Lu 1.03 0.1	Th 6 2.89	U 4.09	Pb 200.74	Ag 1.678	Cr 130.64
128L 127L	58 66	E 1.51 E 0.88	2.14	0.56	0.35	1.06	0.009	0.91 0.69	0.15	2.92 0 3.38 0	0.09 87 0.04 87	.61 1	15.15 3.2 9.15 1.5	6 0.22	38.82 42.42	2.21	9.44 7.62	28.56 19.00	83.56 63.88	1.51 1.16	9.89 9.94	56.68 30.02	3.76	6.96 4.10	2.71 1.64	0.47	0.11	0.49 0. 0.33 0.	34 0.18 25 0.14	0.19 0.0	3 0.71 2 0.42	11.60 16.22	69.30 33.02	0.186	71.10 123.06
126L 125L	74 82	E 0.67 E 1.00	1.80 1.60	0.41 0.47	0.15	0.93	0.005	1.10 0.91	0.15	3.71 (3.80 (0.03 85 0.06 74	.49 1 .10 2	11.95 0.9 21.28 2.5	1 0.11 6 0.17	42.26 87.12	0.77	7.49 3.27	17.17 18.02	62.26 59.52	0.84 1.47	16.12 27.66	26.70	1.41 2.49	2.74 4.84	1.11 1.97	0.19 0.36	0.05	0.23 0. 0.41 0.	18 0.10 33 0.15	0.10 0.0	1 0.28 2 0.40	19.57 73.88	87.74 31.72	0.124	69.59 122.00
124L 123L	90 98	E 1.14 E 1.18	1.96 3.05	1.35 0.95	0.21 0.22	0.76	0.008	0.81	0.29	2.88 (0.04 54 0.05 6.	.03 1 29 2	19.42 2.6 25.10 2.1	8 0.21 5 0.20	118.02 66.70	2 1.66	4.24 9.17	22.34 25.39	74.78 86.17	1.72 2.12	26.53 10.98	34.33 44.69	3.35 3.03	6.32 6.18	2.44 2.61	0.43	0.09	0.43 0. 0.52 0.	35 0.19 43 0.23	0.18 0.0	3 0.58 4 0.54	56.37 23.59	5.85 8.62	0.239	94.60 159.64
122L 121L	120 128	E 2.08 E 1.54	22.57 26.02	1.36 0.57	0.38	0.99	0.013	0.67	0.55	2.23 0	0.02 8.	06 2 .40 1	25.03 5.1 19.44 3.1	6 0.38 7 0.27	72.54	2.90	3.32	96.49 111.41	359.54 428.67	4.10	5.19	100.71 96.46	6.06 4.36	12.07 8.75	5.16 3.67	0.97	0.22	1.02 0. 0.78 0.	80 0.44 61 0.33	0.39 0.0	6 0.97 5 0.70	25.49 18.69	22.86	0.711	191.95 103.37
120L 119L	136	E 2.30 E 0.75	24.04 44.22	0.97	0.42	1.25	0.012	0.68	0.19	1.22 0	0.08 5.	.23 1	23.48 8.0	9 0.33 4 0.14	8.62	3.32	2.45	135.18 192.63	499.48	4.25	2.88	123.27	2.13	4.08	5.00	1.03	0.24	1.07 0. 0.41 0.	88 0.47 32 0.17	0.40 0.0	7 1.02 5 0.65	23.19	24.73	0.909	43.00
117L	163	E 0.54 E 0.41	45.05	0.22	0.08	1.46	0.007	0.35	0.07	1.07 0	0.03 22	.04 2	22.55 1.8	8 0.09	4.66	0.63	13.52	242.51	983.45	0.90	1.65	105.22	1.29	2.03	0.89	0.23	0.07	0.24 0.	16 0.09	0.10 0.0	2 0.36	4.54	3.72	0.210	42.52
115L 114	174	E 0.49	44.18	0.20	0.09	1.43	0.007	0.33	0.09	1.05 0	0.02 15	.68 1	16.93 2.6	6 0.10	5.10	0.60	13.56	181.65	736.25	0.94	1.51	94.11	1.41	2.88	1.25	0.24	0.06	0.25 0.	21 0.09	0.09 0.0	1 0.28 2 0.30	3.46	2.15	0.129	40.37
113L 112L	190 198	E 0.80 E 0.69	43.09 43.10	0.36	0.14	1.39	0.008	0.37	0.07	1.10 0	0.03 21	.17 1	19.00 1.1 10.53 2.9	1 0.14	7.32	1.12	15.84 11.25	216.87 194.90	870.19 776.86	1.74	1.14	96.74 88.64	2.24	4.68	1.93	0.36	0.10	0.36 0.	36 0.19 29 0.15	0.15 0.0	3 0.42 2 0.36	4.02	5.28 4.90	0.120	55.35 53.34
111L 110L	234 242	E 0.73 E 0.61	43.58 44.49	0.40	0.13	1.36	0.010 0.008	0.41	0.08	1.11 (0.04 21 0.02 15	.30 1 .59 1	18.41 2.3 14.49 2.5	i2 0.12	8.99 5.85	0.96	14.19 11.99	258.41 226.73	1040.35 916.28	1.56	1.20 0.99	102.14 93.08	2.06	4.26 3.28	1.80 1.34	0.41 0.26	0.10	0.38 0. 0.28 0.	29 0.15 23 0.12	0.16 0.0	2 0.38 2 0.29	4.95 3.58	12.08	0.179	49.58 53.11
109L 108L	250 258	E 1.08 E 2.06	44.08 42.64	0.58	0.20	1.51	0.010	0.34	0.06	1.25 (1.66 (0.05 37 0.09 17	.17 1 .51 3	13.45 3.5 31.06 8.3	i9 0.21	10.95 19.78	1.52	13.26 18.00	176.21 183.78	693.93 700.52	2.91 4.98	1.50	94.28 105.94	3.90 6.69	7.98 14.26	3.34 5.84	0.68	0.18	0.70 0. 1.24 1.	60 0.31 04 0.50	0.27 0.0	7 1.10 8 1.43	2.94 2.92	9.89 15.43	0.269	97.00 135.37
107L 106L	266 274	E 2.22 E 2.69	42.23 39.70	1.33	0.36	1.61	0.021 0.030	0.44	0.08	1.89 0	0.09 21 0.11 22	.12 2	26.53 13. 29.03 13.	68 0.42 97 0.43	19.96	3.28	7.20 9.03	196.99 234.90	759.74 899.95	5.71 6.52	1.29	106.89	9.14	17.00 19.21	6.91 7.95	1.40 1.59	0.31	1.47 1. 1.63 1.	19 0.61 36 0.70	0.59 0.0	9 1.59 0 1.85	3.53 4.73	14.99 11.34	0.340	124.42
105L 104L	282 290	E 5.34	35.08	2.79	0.76	1.48	0.053	0.68	0.12	2.93 0	0.14 22	.06 2	25.37 22. 35.32 24.	38 0.76 01 0.87	45.24	7.00	4.57	332.65	1288.72	11.26	3.10	174.75	15.32	30.98	13.17	3.01	0.61	2.73 2.	23 1.10 68 1.33 74 4.00	0.99 0.1	7 3.01 9 3.44	9.15	24.20	0.597	136.95
102L	290 342 348	D 13.80	9.32	7.62	2.50	3.30	0.063	1.21	0.09	4.89 0	0.56 13	.10 2	45.91 71. 71.00 75	29 2.35 62 2.22	140.88	3 22.55	4.47	194.76 235.63	433.30	27.37	12.68	357.76	43.05	84.84 82.06	37.40	7.45	1.64	4.50 3. 7.63 6. 7.73 6	13 3.06 19 3.10	2.69 0.4	2 8.47	7.65	75.90	1.437	264.33
100L	356	D 15.4	8.46	8.10	2.66	3.52	0.060	1.35	0.22	5.09 0	0.61 15	.79 7	72.21 76.	37 2.36	148.11	23.23	6.22	206.11	455.83	27.68	24.70	364.23	44.58	83.62	38.69	7.81	1.68	7.85 6.	19 3.08	2.74 0.4	4 8.67	7.88	81.03 101.94	1.633	261.41
98L 97L	372 380	D 10.53	10.23	7.07	2.07	3.07	0.059	1.49	0.18	4.78 (0.29 21	.43 5	56.90 56. 53.27 42.	70 2.05 70 1.50	148.48	3 19.82 9 14.78	3.90	202.17 557.84	524.98 2188.98	26.73	66.45 73.50	306.27	38.68	78.98	33.56 25.83	6.86 5.15	1.49	7.06 5. 5.49 4.	70 2.91 48 2.35	2.63 0.4	4 7.54 4 5.78	16.85	169.66 136.33	2.296	242.69
96L 95L	388 396	D 5.88 D 8.39	41.56 29.64	3.47 4.78	0.94	1.50 2.05	0.028	0.96	0.24	3.39 (4.18 (0.13 18 0.22 20	.81 3 .18 4	35.61 30. 45.59 41.	71 1.04 58 1.43	74.78	9.15 3 13.21	7.52 4.55	1105.43 797.59	4623.97 3190.14	15.29 20.30	50.05 74.72	193.91 231.07	20.20	41.95 56.16	17.70 23.23	3.42 4.75	0.76	3.69 3. 4.91 4.	08 1.64 06 2.11	1.47 0.2 1.98 0.3	4 3.70 2 5.23	20.03 22.73	77.45 92.94	1.575 1.513	103.72 144.44
94L 93L	441 449	D 8.45	28.94 27.47	4.99 5.05	1.33	2.13	0.040	1.03	0.19	4.02 0	0.14 21 0.13 21	.98 4 .97 2	43.22 38. 29.97 35.	06 1.37 48 1.40	107.50	13.59 12.84	9.29 10.31	681.13 382.79	2680.54 1417.27	20.17	62.14 47.19	238.44	28.00	57.34 54.05	24.24 22.34	4.89 4.46	1.05	5.05 4. 4.64 3.	.08 2.08 .81 2.03	1.88 0.3 1.85 0.3	2 5.41 0 4.83	19.34 16.49	111.91 117.73	2.070 2.567	153.70 162.71
92L 91L	457	D 10.38	15.80 35.42	6.34 4.12	1.64	2.79	0.052	1.43	0.20	4.11 (3.43 (0.15 24 0.15 21	.15 5	55.00 54. 39.70 29.	39 1.68 16 1.15	163.22	10.95	3.96	222.94	661.51 2972.35	24.92	110.04	4 273.15	23.64	67.53 46.16	27.76	5.59	0.86	5.88 4. 4.02 3.	74 2.58 22 1.69	2.38 0.3	9 6.53 3 4.49	29.80	164.69	3.710	191.27
90L 89L	468	D 3.34	48.60 35.40	3.95	0.53	1.00	0.017	1.14	0.25	3.55 0	0.14 18	.86 4	27.34 15. 18.16 28.	19 0.60 44 1.20	48.03	10.64	11.63	12/4.51 930.49	5435.99 3821.69	9.07	39.26	262.26	22.64	23.78 46.13 21.47	9.90	1.97 3.76	0.91	2.10 1. 3.98 3.	34 1.74	1.64 0.2	4 2.06 9 4.89 2 1.06	15.86	73.49 88.14	1.551	128.99
87L 86I	490	D 7.48	25.17	4.25	1.13	1.77	0.035	1.35	0.23	4.27 0	0.12 19	.22 4	19.35 30. 54 17 59	56 1.18	104.42	2 11.39	19.08	667.57	2673.63	19.26	87.63 109.73	392.41	28.37	54.93 83.84	22.93 37.91	4.57	1.06	4.77 3.	99 2.08 23 3.31	1.84 0.3	0 5.18	22.16	227.78	4.666	135.63
85L 84L	504 512	D 7.65 D 6.24	21.54 34.32	4.99	1.50	2.08	0.041 0.028	1.25	0.18	4.22 (0.14 24 0.09 4.	.77 5 56 3	50.75 40. 39.08 26.	52 1.41 47 0.99	112.38	3 14.39 9.57	8.75	499.70 972.82	1912.70 4048.41	20.22	78.47	269.29	29.23	59.90 43.81	24.80 18.21	5.04 3.65	1.12	5.13 4. 3.78 3.	18 2.19	2.00 0.3	3 5.55 2 3.93	18.80 18.95	143.03 134.80	2.756	165.81 108.79
83L 82L	519 527	C 18.11 C 19.01	3.72	8.43 8.79	3.30 3.55	2.63 2.78	0.087	1.60 1.68	0.09	4.77 (4.09 (0.68 3. 0.84 4.	99 5 57 6	59.18 74. 54.94 78.	73 2.89 13 3.14	183.84	1 31.34 1 34.31	18.38 27.53	202.99 186.20	353.25 247.64	44.18 51.55	28.87 5.88	537.21 733.31	69.53 73.77	134.93 143.92	61.79 67.45	12.26 13.68	2.59 1 2.81 1	2.54 9. 3.58 10	.67 4.69 .86 5.59	4.09 0.6 4.87 0.8	4 12.47 0 13.24	8.84 6.36	67.65 26.65	0.812	158.86 154.88
81L 80L	535 543	C 19.3	1.85	9.06 10.55	3.50 3.49	2.82	0.096	1.73 1.64	0.13	4.02 0	0.96 7. 0.92 5.	01 5 23 5	59.18 59. 56.33 82.	45 2.98 14 3.17	193.74	4 33.45 3 34.29	17.89 19.68	175.49 172.53	218.54 200.17	46.66 52.11	3.87 3.36	753.91	73.44	144.33 148.37	67.22 68.76	13.40 13.94	2.79 1 2.94 1	3.67 10 3.95 10	.53 5.18 .96 5.50	4.54 0.7 4.71 0.7	3 13.17 5 13.37	6.65 6.88	24.76 26.19	0.491 0.528	147.80 155.32
79L 78L	548 554	C 18.54	1.51	7.62	3.61	2.80	0.090	1.88	0.16	0.21 0	0.75 3. 0.90 5.	72 6 07 6	58.34 58. 58.27 77.	76 3.13 89 3.40	218.23	5 34.45 3 35.65	29.76	172.66	198.33	47.18	1.78	816.66	5 71.66 2 75.97	139.93 148.63	65.18 68.69	13.25	2.72 1	3.07 10	.26 5.03	4.55 0.7	5 13.05 9 13.47	6.79 7.20	23.60	0.372	158.86
77L 76I	570	C 19.14	1.44	8.79	3.59	2.82	0.087	1.79	0.15	0.07 0	0.82 5. 0.97 5.	59 5 44 5	56.73 68. 55.33 63	79 2.98 92 3.11 89 2.90	218.97	35.75	30.70	173.22	192.92	42.19	1.85	800.02	76.16	120.50	59.23 69.53	14.07	2.86 1	4.19 11 2.07 0	07 4.35	4.83 0.7	7 13.29	6.20 6.32	25.23	0.502	163.57
75L 74L	590 597	C 20.9	1.15	8.78	3.72	2.97	0.104	1.71	0.18	0.06 0	0.83 6.	53 4 21 5	40.42 73. 56.39 70.	64 3.15 24 2.15	195.06	6 36.02 33.94	23.12	172.05	172.07	47.88	0.72	784.20	0 74.80 6 68.77	147.79	67.55	13.53	2.77 1	3.48 10	37 5.16	4.35 0.7	0 13.07	6.33	24.86	0.379	163.92
73L 72L	613 621	C 14.8 C 19.4	1.98 3.03	8.95 10.05	3.39 3.21	2.60 2.90	0.100	1.76 1.42	0.16	0.12 0	0.48 8. 0.71 8.	33 4 48 6	42.46 60. 64.69 70.	92 2.00 23 2.05	147.75	5 31.42 3 32.22	8.81 5.14	166.04 186.09	241.21 304.10	44.94 54.89	1.68 5.50	788.45	62.25 73.15	121.88 144.90	60.06 69.96	12.41 14.22	2.62 1 3.03 1	2.14 9. 4.34 11	68 4.88 .74 5.94	4.30 0.6 5.10 0.7	0 10.15 7 12.81	5.74 7.56	26.12 39.84	0.240	140.66 152.84
71L 70L	627 633	C 21.10	2.61	9.34 8.50	3.32 3.35	2.81 2.67	0.122 0.111	1.54 1.54	0.22	0.38 0	0.95 8. 0.92 11	91 4 .20 6	49.75 82. 52.12 73.	02 2.14 37 2.06	208.53	3 36.31 35.07	9.74 11.90	193.21 177.65	288.02 241.48	72.99 65.78	5.60 5.01	746.15	92.21 82.44	190.26 176.31	94.94 84.70	19.64 17.69	3.90 1 3.55 1	9.62 16 7.77 14	.01 7.83	6.75 1.0 6.11 0.8	0 16.86 9 15.43	9.65 9.50	38.03 36.39	0.577	140.50 135.91
69L 68L	641 649	C 21.29	1.97	8.56	3.52	2.70	0.120	2.32	0.23	0.20 0	0.90 11 0.85 9.	.15 4 48 3	13.17 43. 36.50 74.	94 3.13 26 3.07	181.79	9 34.48 7 34.15	10.86	158.49	187.01	64.86 69.67	3.01	724.49	22.77	47.03 50.06	87.95 95.37	17.93	3.82 1	8.00 14 9.54 15	.06 6.80	5.62 0.9	1 16.99 6 16.74	8.84 8.48	32.58	0.582	122.04
66L	664	B 19.99	1.77	16.38	3.07	2.59	0.222	2.61	0.36	0.09 0	0.63 9.	48 4	19.15 70. 38.42 70.	39 3.00 97 3.10	173.59	30.64	8.20	133.04	137.80	51.61	2.54	670.38	16.98	35.90 43.78	63.19 80.93	12.84	3.01 1	6.22 12	.56 5.32	4.55 0.7	6 12.42 8 15.83	6.31 7.79	33.60	0.471	123.40
64L 631	678	B 21.52	1.25	9.49 9.67 5.76	3.38	2.83	0.141	2.43	0.22	0.11 0).82 5.).91 4	70 4 53 3 08 3	4.03 52. 37 31 26	49 3.26	176.10	34.65	6.92	142.68	109.30	57.40 59.50	3.29	656.09	22.75	46.41	85.05 44 11	14.11	3.69 1	4.36 12 7.02 12 8.67 6	.74 6.02	5.42 0.8	5 17.40 7 7.88	7.56	41.64	0.557	119.54
62L 61L	692 707	B 19.49 B 20.09	1.46	7.99	3.50	2.85	0.096	3.19	0.23	0.09 0	0.90 4. 0.94 6.	52 3 06 3	39.00 35. 34.47 35.	54 2.99 80 2.90	170.83	8 29.75 8 30.38	4.99	144.55	175.28	38.37 35.93	2.42	752.87	14.71	30.21 29.43	56.01 54.87	11.18 11.03	2.60 1 2.57 1	0.99 8.	19 3.94 81 3.72	3.33 0.5 3.14 0.5	4 10.30 6 10.37	4.47	24.49 25.50	0.470	146.16
60L 59L	715 723	B 18.95 B 20.23	1.52	6.44 11.62	3.45 3.62	2.68 2.97	0.063 0.147	3.59 2.88	0.19 0.30	0.10 0).91 5.).85 7.	44 2 14 4	20.98 31. 14.32 38.	78 2.65 97 2.83	156.68	8 28.05 5 30.05	3.53 7.11	138.99 147.03	192.41 177.73	34.92 42.52	1.48 1.94	699.86 755.76	i 13.02	27.41 31.19	50.78 57.43	10.13 11.71	2.36	9.84 7. 1.59 8.	44 3.64 91 4.32	3.08 0.5 3.70 0.6	1 9.43 1 10.90	4.71 5.44	23.57 26.12	0.340	128.37 137.49
58L 57L	731 739	B 20.61	1.69	8.34 10.41	3.74	4.11 3.39	0.115	2.92	0.18	0.14 0	0.90 5. 0.85 5.	62 3 57 4	38.92 39. 14.13 38.	49 2.92 17 3.03	185.28	30.36 30.18	4.58	143.52	166.60	39.67	1.98	698.94	14.54	29.85 30.01	54.62 55.06	11.01	2.57 1	0.72 8.	30 4.11 14 3.97	3.56 0.5	8 10.46 6 10.24	4.60 5.52	27.17 26.06	0.429	244.58
56L 55L	755	B 19.73 B 20.14	1.34	9.05	3.58	2.90	0.106	2.98	0.21	0.08 0).90 6.).88 7.	09 4 33 4	17.71 42.	59 2.92 39 3.17	174.84	30.42 31.88	6.43	145.50	1/1./9	39.53 43.25	1.4/	755.87	14.68	30.28	56.65 61.06	11.31	2.63 1	1.29 8. 2.09 9.	39 4.05 24 4.41	3.49 0.5	7 10.41 4 11.47	5.32	26.03	0.438	146.70
52L 511	779 787	B 21.44	0.94	10.47	3.40	3.01	0.166	2.54	0.22	0.16 0	0.81 6.	09 5	50.30 81. 50.95 44	43 3.37 67 3.03	195.25	5 33.75 32.00	10.43	144.60	134.12	43.49 51.31 44.48	4.17	746.30	17.58	36.35	66.65	13.57	3.16 1	3.49 10 2.23 9	78 5.18	4.35 0.7	8 13.01 3 11.98	5.81	30.03	0.528	166.46
49L 48L	803 811	B 20.50 B 18.73	1.41	8.19 6.84	3.65 3.46	3.01 2.68	0.099 0.076	2.95 3.33	0.18	0.12 0	0.93 4. 0.90 4.	63 4 29 3	19.55 41. 37.50 34.	72 3.11 71 3.07	184.02	2 31.94	10.12 5.21	150.61 143.61	178.41 180.34	43.63 35.81	2.00	760.68	15.94 13.30	32.78 27.57	61.31 51.20	12.39 10.26	2.78 1 2.39 1	2.15 9. 0.19 7.	27 4.49 64 3.73	3.87 0.6 3.15 0.5	2 11.46 2 9.48	5.45 4.32	26.87 24.83	0.420	145.61 128.74
46L 44L	827 846	B 20.63	1.18 1.45	8.02 9.87	3.71 3.59	2.97 2.81	0.101 0.159	3.08 2.84	0.18	0.08 0).94 5.).88 5.	45 3 45 3	34.14 41. 39.42 40.	15 3.04 66 3.13	184.05	5 31.75 5 30.51	4.68 6.00	148.95 147.41	164.28 173.89	40.20 38.80	2.52	746.68	15.14 14.71	31.62 30.44	58.22 55.55	11.85 11.27	2.66 1	1.57 8. 0.86 8.	71 4.07 24 4.02	3.50 0.6 3.44 0.5	0 10.88 9 10.64	5.43 5.05	25.53 27.07	0.412 0.479	144.40 132.73
42L 40L	862 878	B 19.43 B 20.01	1.36	8.55 8.61	3.53	2.89	0.100	3.20	0.20	0.07 0).93 5.).93 6.	05 3 57 2	35.96 37. 26.84 41.	65 3.13 37 3.17	169.22	2 30.13 3 31.22	5.03	143.19 148.71	174.37 173.55	40.07	2.50	735.36	14.92 15.33	30.85 31.58	57.56 58.50	11.55	2.67 1	1.43 8. 1.55 8.	48 4.18 76 4.25	3.64 0.6 3.80 0.6	7 10.70 2 10.96	5.19 5.17	24.34	0.519	135.55 136.56
38L 37L	904 912	в 19.48 В 18.53 В 10.74	1.17	9.62 8.84	3.44	2.84	0.108	1.96	0.19	0.08 0).85 5.).87 4.	7U 4 43 7 38 7	44.37 44. 70.71 40. 76.96 47	90 3.19 42 3.40 80 £ 1.4	170.03	31.65 330.93	7.40 8.02	147.82	158.74	43.99	4.31	762.80	15.47	33.22 32.12	61.78 58.56 58.07	12.57	2.85 1	2.36 9. 1.92 9. 2.12 0	37 4.53 37 4.78	3.81 0.6 4.34 0.5	0 12.11 9 11.24 3 11 44	5.67 4.80	23.76	0.381	141.21
35L 33L	920	B 18.45	1.22	8.08	3.36	2.82	0.079	1.90	0.19	0.04 0	0.88 7. 0.88 4	11 4 34 6	45.14 41. 61.25 67	62 4.47 39 2.93	176.48	3 31.06	7.05	151.53	176.87	46.23	0.70	743.66	15.69	32.48	59.69 63.05	11.99	2.88 1	2.08 9.	49 4.78	4.28 0.5	7 11.42	5.25	23.61	0.411	141.69
32L 31L	944 952	B 19.98	1.17	10.07 7.88	3.55 3.48	2.88 2.98	0.133	1.59	0.22	0.08 0	0.83 5. 0.93 6.	18 9 27 6	96.61 72. 54.81 40.	58 5.92 08 3.37	183.76	6 33.13 7 31.03	8.16 8.89	157.01 154.93	176.84 190.02	49.09 44.38	1.40	760.64	17.02	35.43 31.35	62.82 57.29	12.62 11.57	2.99 1 2.86 1	2.79 10 1.57 9.	.02 5.14	4.65 0.6	4 12.70 1 10.94	6.68 5.29	29.29 23.65	0.488	146.32 149.68
29L 27L	969 981	B 19.2	1.38	9.41 8.65	3.55 3.32	3.03 3.10	0.108	1.84	0.23	0.08 (0.90 5. 0.92 4.	01 4 83 7	47.64 65. 74.22 56.	83 2.60 69 3.35	182.17	7 31.65 3 30.09	11.55	151.78 147.64	175.95 175.97	46.75 43.76	0.97	781.94	16.16	33.05 30.58	61.02 55.22	12.35 11.16	2.98 1 2.75 1	2.36 9. 1.19 8.	69 4.86 89 4.57	4.19 0.5 4.02 0.5	9 11.85 7 10.93	4.82 6.55	26.13 33.44	0.431	144.59 170.49
25L 24L	996 1003	B 19.0	1.17	8.38 8.31	3.56	3.04	0.087	1.84	0.21	0.04 0	0.88 4.	70 5 18 5	54.66 48. 57.29 41.	40 4.22	174.61	31.74	8.65	154.64 152.87	182.34	44.94	0.59	770.79	15.30	32.22 31.30	58.63 57.76	11.85	2.90 1	2.09 9.	29 4.66 25 4.69	4.01 0.5	7 11.35	5.46 4.96	23.12	0.401	143.71
22L 20L	1017 1033	B 19.39	1.21	8.72 9.57	3.60	3.13	0.090	1.96	0.22	0.05 0	0.86 6. 0.88 7.	46 5 10 7	59.50 49. 78.84 69. 75.42 74	2/ 2.66	174.20	2 33.30	8.16	153.98	182.71	44.80	0.75	773.59	15.60	32.03	59.56 61.26	12.02	2.91 1 3.02 1	2.05 9.	35 4.70 .04 5.18	4.09 0.5	/ 11.49 7 12.08	5.41 5.60	23.02 25.56	0.390	148.29
16L 14I	1049 1065 1081	B 19.8	1.53	9.05 10.19 10.47	3.62	4.04	0.100	1.61	0.20	0.07 0	0.85 6. 0.58 7	20 / 57 6 56 8	51.06 76. 32.59 87	32 3.37 12 3.72	195.52	2 32.97	8.78	155.26	169.11 169.27 239.42	49.27	1.04	701.10	16.08	33.23 30.76	60.22 54 76	12.39	3.06 1	2.41 10	.08 5.09 44 4.88	4.54 0.6	5 12.16	4.95	26.66 57.56	0.410	208.53
13L 12L	1089	A 17.53	4.41	10.27	2.88	3.63	0.046	2.18	0.23	4.85 (0.67 3.	69 7 22 7	78.69 83.	00 2.75	163.96	6 27.84 1 26.28	10.18	204.89	508.25	45.15	7.24	543.20	14.77	30.70	54.36 50.85	11.15	2.72 1	1.32 9. 0.68 8.	28 4.76	4.19 0.6	1 11.47	5.96	56.69 89.02	0.946	230.22
11L 10L	1120 1128	A 17.15	5.79 2.85	9.40 10.41	2.97	3.43 3.27	0.073	1.81 2.13	0.26	2.89 (4.72 (0.74 7. 0.76 7.	91 5 60 6	59.20 72. 52.00 80.	73 4.35 58 4.07	168.18 178.49	3 27.78 9 29.71	8.28 7.52	205.76 161.81	494.36 265.62	43.52 46.74	5.86 8.29	605.24 557.13	14.35	29.58 31.33	53.09 56.92	10.85 11.58	2.64 1 2.81 1	1.05 8. 1.62 9.	71 4.53 58 4.88	3.97 0.5 4.26 0.6	6 10.97 3 11.82	4.94 6.31	46.77 67.20	0.866	202.19 191.56
9L 7L	1135 1150	A 18.05	2.75	11.90 11.15	3.00 3.13	3.30 3.21	0.497	1.93 1.79	0.48	2.92 ().72 7.).75 8.	77 8 13 8	81.75 81. 31.67 87.	32 2.40 85 3.21	184.87	7 30.32 3 31.68	10.57 9.99	169.40 122.23	278.20	52.05 47.84	5.18 4.73	528.23 538.51	15.68	32.55 32.50	57.76 59.60	11.86 12.11	2.91 1 2.93 1	2.22 10	.19 5.48	4.93 0.6 4.59 0.6	8 12.07 7 12.14	6.54 5.35	56.01 55.00	0.953	161.29 168.15
5L 3L	1166 1182	A 18.65	1.65	12.93	3.05	3.54	0.280	1.89	0.34	2.27 0	0.72 8.	62 7 80 7	72.72 89.	13 3.93 19 5.01	188.89	30.93	13.68	149.74	181.49	55.77	3.71	543.00	16.32	34.68	61.42 59.82	12.72	3.09 1	2.97 11	.08 5.90	5.41 0.7	/ 12.47 2 12.07	5.86	45.15	0.793	166.27
2L 1L	1190	A 18.80	2.18	10.97	3.17 2.96	3.41 4.28	0.054	2.04	0.22	4.39 ().76 8.).68 9.	ວບ 6 20 6	56.17 86.	od 3.32 75 2.11	183.33	3 30.89 3 29.35	10.13	142.54	157.51 210.66	49.75	10.11 6.22	525.19 486.16	15.85	32.91 31.32	59.63 56.83	12.12	2.98 1	∠.49 10 1.81 9.	. 12 5.11 59 4.94	4.53 0.6	+ 12.50 6 12.08	ь.12 5.18	db.31 38.83	2.090	234.86

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Table 32	Lab code	Unit	Denth (cm)	²⁰⁶ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁴ Pb	²⁰⁸ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁶ Pb	²⁰⁸ Pb/ ²⁰⁶ Pb	T (Ma)	ш	к
Leachate	129L	E	50	18.677	15.675	38.777	0.83930	2.07610	108.9	9.750	3.940
Leachate	128L	E	58	17.988	15.606	37.854	0.86760	2.10440	511.8	9.700	3.890
Leachate	127L 126L	F	66 74	18.053	15.610	37.930	0.86470	2.10100	470.8	9.700	3.890
Leachate	125L	Ē	82	17.928	15.596	37.756	0.86990	2.10590	541.5	9.690	3.870
Leachate	124L	E	90	18.360	15.643	38.361	0.85200	2.08940	294.6	9.730	3.920
Leachate	123L	E	120	18.315	15.638	38.306	0.85390	2.08620	320.6	9.730	3.930
Leachate	121L	E	128	18.522	15.662	38.606	0.84560	2.08430	203.0	9.740	3.940
Leachate	120L	E	136	18.464	15.663	38.542	0.84830	2.08740	244.9	9.750	3.950
Leachate	118L	E	152	18.299	15.642	38.297	0.85480	2.09280	336.4	9.730	3.920
Leachate	117L	E	163	18.238	15.637	38.209	0.85740	2.09510	373.4	9.730	3.920
Leachate	116L 115L	F	168 174	18.457	15.6657	38.518	0.84880	2.08700	253.4	9.760	3.940
Leachate	114L	E	182	18.503	15.661	38.574	0.84640	2.08480	215.8	9.750	3.940
Leachate	113L	E	190	18.327	15.644	38.328	0.85360	2.09140	318.9	9.730	3.920
Leachate	112L 111L	E	234	18.578	15.645	38.682	0.84350	2.09250	174.6	9.740	3.950
Leachate	110L	E	242	18.581	15.670	38.684	0.84340	2.08180	171.4	9.760	3.950
Leachate	109L 108l	E	250 258	18.375	15.646	38.395 38.510	0.85150	2.08950	288.1	9.730	3.930
Leachate	107L	E	266	18.565	15.666	38.660	0.84380	2.08240	177.2	9.750	3.940
Leachate	106L	E	274	18.634	15.673	38.754	0.84110	2.07990	137.2	9.750	3.950
Leachate	103L	E	290	18.684	15.680	38.816	0.83920	2.07820	109.7	9.760	3.950
Leachate	103L	E	298	18.684	15.680	38.802	0.83920	2.07670	110.4	9.760	3.940
Leachate	102L 101L	D	342	18.662	15.677	38.793	0.83980	2.07820	118.8	9.760	3.950
Leachate	100L	D	356	18.656	15.674	38.775	0.84020	2.07850	122.8	9.750	3.950
Leachate	99L	D	364	18.661	15.675	38.783	0.84000	2.07820	120.7	9.760	3.950
Leachate	97L	D	380	18.673	15.681	38.805	0.83980	2.07830	119.7	9.770	3.950
Leachate	96L	D	388	18.693	15.681	38.824	0.83880	2.07690	104.7	9.760	3.950
Leachate	95L 94I	D	396 441	18.679	15.680	38.799	0.83940	2.07730	114.2	9.760	3.940
Leachate	93L	D	449	18.664	15.678	38.787	0.84000	2.07810	122.1	9.760	3.950
Leachate	92L	D	457	18.681	15.678	38.804	0.83920	2.07710	109.9	9.760	3.950
Leachate	91L 90L	D	462	18.685	15.677	38.802	0.83920	2.07650	107.8	9.760	3.940 3.940
Leachate	89L	D	474	18.647	15.674	38.782	0.84060	2.07990	128.9	9.760	3.950
Leachate	88L	D	482	18.642	15.675	38.786	0.84088	2.08064	134.1	9.760	3.960
Leachate	86L	D	498	18.630	15.671	38.756	0.84113	2.08025	136.8	9.750	3.950
Leachate	85L	D	504	18.657	15.678	38.781	0.84033	2.07865	126.8	9.760	3.950
Leachate	83L	C	512	18.607	15.673	38.768	0.84087	2.08044	131.8	9.760	3.950
Leachate	82L	С	527	18.703	15.685	38.892	0.83861	2.07938	102.3	9.770	3.970
Leachate	81L 801	C	535 543	18.713	15.686	38.910 38.962	0.83821	2.07920	96.6 80.7	9.770	3.980
Leachate	79L	C	548	18.751	15.697	38.981	0.83714	2.07888	83.5	9.790	3.990
Leachate	78L	С	554	18.753	15.695	38.971	0.83688	2.07811	78.8	9.790	3.980
Leachate	2L 77L	C	562	18.760	15.696	38.990	0.83666	2.07828	75.2	9.790	3.990
Leachate	76L	Č	578	18.759	15.696	38.989	0.83673	2.07853	76.4	9.790	3.990
Leachate	75L	C	590	18.744	15.693	38.969	0.83719	2.07904	82.5	9.780	3.980
Leachate	74L 73L	C	613	18.735	15.691	38.947	0.83753	2.07884	87.4	9.780	3.980
Leachate	72L	С	621	18.714	15.685	38.896	0.83814	2.07843	94.9	9.770	3.970
Leachate	71L 70L	C C	627	18.733	15.689	38.919	0.83751	2.07763	86.3 97.8	9.780	3.970
Leachate	69L	C	641	18.744	15.688	38.962	0.83698	2.07867	77.4	9.770	3.980
Leachate	68L	C	649	18.756	15.685	38.919	0.83627	2.07504	64.9	9.770	3.960
Leachate	66L	B	664	18.740	15.684	38.918	0.83675	2.07636	72.2	9.770	3.960
Leachate	65L	В	670	18.780	15.691	38.945	0.83553	2.07372	55.6	9.780	3.950
Leachate	64L 63L	B	678 686	18.756 18.743	15.698 15.689	38.970 38.960	0.83699	2.07775 2.07854	81.9 78.7	9.790 9.770	3.980
Leachate	62L	В	692	18.748	15.694	38.961	0.83709	2.07814	81.4	9.780	3.980
Leachate	61L	B	707	18.729	15.691	38.935	0.83782	2.07885	92.0	9.780	3.980
Leachate	59L	B	715	18.746	15.695	38.961	0.83720	2.07830	83.6	9.790	3.980
Leachate	58L	В	731	18.738	15.690	38.951	0.83736	2.07871	84.4	9.780	3.980
Leachate	57L 56L	B	739	18.735	15.693	38.953	0.83761	2.07915	89.4 78.3	9.780	3.980
Leachate	55L	В	755	18.745	15.690	38.954	0.83710	2.07810	79.8	9.780	3.980
Leachate	53L	B	771	18.759	15.689	38.961	0.83630	2.07690	67.8	9.770	3.970
Leachate	51L	B	787	18.754	15.692	38.961	0.83670	2.07900	75.9	9.780	3.980
Leachate	49L	В	803	18.728	15.690	38.931	0.83777	2.07870	90.7	9.780	3.980
Leachate	48L 46L	B	811 827	18.764	15.696	38.920	0.83810	2.07930	95.5 72.8	9.780	3.980
Leachate	44L	В	846	18.753	15.697	38.986	0.83700	2.07880	82.0	9.790	3.990
Leachate	42L 401	B	862 878	18.754	15.696	38.990 38.984	0.83690	2.07890	79.5	9.790	3.990
Leachate	38L	В	894	18.741	15.691	38.960	0.83720	2.07880	82.7	9.780	3.980
Leachate	37L	B	904 012	18.701	15.690	38.899	0.83900	2.08020	109.9	9.780	3.980
Leachate	35L	В	920	18.754	15.696	38.979	0.83690	2.07850	79.9	9.790	3.980
Leachate	33L	В	936	18.721	15.691	38.939	0.83820	2.07990	98.0	9.780	3.980
Leachate	32L 31L	B	944 952	18.734	15.690	38.946	0.83820	2.08060	98.1 86.6	9.780 9.780	3.990
Leachate	29L	В	969	18.754	15.692	38.959	0.83670	2.07740	75.4	9.780	3.980
Leachate	27L	B	981 00e	18.742	15.687	38.932	0.83700	2.07730	77.9	9.770	3.970
Leachate	24L	В	1003	18.739	15.689	38.946	0.83730	2.07840	82.7	9.780	3.980
Leachate	22L	В	1017	18.752	15.691	38.970	0.83670	2.07820	75.1	9.780	3.980
Leachate	20L 18L	B	1033	18.747	15.692	38.967 38.979	0.83700	2.07850	79.5 76.2	9.780 9.780	3.980
Leachate	16L	В	1065	18.735	15.693	38.952	0.83760	2.07910	89.5	9.780	3.980
Leachate	14L	A	1081	18.737	15.685	38.916	0.83710	2.07690	79.1	9.770	3.960
Leachate	12L	A	1098	18.720	15.678	38.873	0.83750	2.07660	82.6	9.760	3.950
Leachate	11L	A	1120	18.723	15.685	38.902	0.83770	2.07770	88.1	9.770	3.970
Leachate	9L	A	1128	18.735	15.686	38.912	0.83720	2.07690	79.1 85.1	9.770	3.960
Leachate	7L	A	1150	18.741	15.687	38.930	0.83700	2.07720	77.5	9.770	3.970
Leachate	5L 31	A	1166	18.748	15.688	38.938 38.931	0.83680	2.07700	74.6	9.770	3.970
Leachate	2L	A	1190	18.747	15.686	38.917	0.83670	2.07600	72.6	9.770	3.960
Leachate	1L	A	1198	18.734	15.686	38.919	0.83730	2.07740	82.3	9.770	3.970
Residue	104 K	D	290	18.753	15.691	38.959	0.83650	2.07680	70.0	9.779	3.970
Residue	98 R	D	372	18.774	15.698	38.973	0.83610	2.07580	67.7	9.789	3.971
Residue	94 R	D	441	18.733	15.684	38.892	0.83730	2.07620	80.8	9.767	3.956
Residue	89 R	D	474	18.724	15.681	38.881	0.83750	2.07660	83.3	9.762	3.956
Residue	77 R	C	570	18.742	15.689	38.973	0.83710	2.07950	80.4	9.776	3.987
Residue	69 R 61 R	C B	641 707	18.728 18.744	15.688	38.909	0.83760	2.07770	88.5 85.7	9.775 9.787	3.967
Residue	52 R	В	779	18.793	15.699	39.000	0.83530	2.07520	55.8	9.790	3.972
Residue	43 R	В	851.5	18.748	15.693	38.985	0.83710	2.07950	81.1	9.783	3.989
Residue	35 K 26 R	B	920	18.736	15.694	38.986	0.83760	2.08070	90.6	9.789	3.996
Residue	17 R	В	1057	18.776	15.694	38.993	0.83590	2.07670	61.8	9.782	3.977
Residue	10 R	Α	1128	18.787	15.698	39.025	0.83550	2.07710	58.1	9.788	3.986

Depth (cm)	Laboratory code	Material	δ ¹³ C	¹⁴ C age (BP)	Calendar age (BC-AD) (2 σ)
186	Lyon-9957 (SacA 32583)	Vegetal matter	-26.6	665 ± 30	1276 -1391 AD
362	Lyon-9959 (SacA 32584)	Wood	-30.8	1455 ± 30	558 - 649 AD
460	Lyon-9960 (SacA 32585)	Seeds	-31.3	2020 ± 30	106 BC - 58 AD
680-690	UGAMS (17652)	Pollen	-27.8	2000 ± 40	111 BC - 83 AD
1080-1090	UGAMS (17352)	Seeds	-25.9	1990 ± 25	43 BC - 63 AD
1189	Lyon-9962 (SacA 32587)	Vegetal matter	-31.9	2040 ± 30	163 BC - 47 AD

Table S3