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Pollutant lead reveals the pre-Hellenistic occupation and ancient growth of Alexandria, Egypt

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[1] It is generally accepted that Alexandria ad Aegyptum was founded ex nihilo in 331 BC by Alexander the Great, rapidly growing into one of antiquity's most opulent economic and intellectual centers. However, ancient texts by Strabo (17.1.6) and Pliny (NH 5.11.62) suggest the existence of a pre-Hellenistic settlement named Rhakotis. This literary evidence has fuelled contentious scholarly debate for decades. Here we present new geochemical data from Alexandria's ancient bay sediments, elucidating unequivocal proof for pollutant lead (Pb) input into the harbor during the Egyptian Old Kingdom (2686–2181 BC). A second contamination peak is detected during the Iron Age (1000–800 BC), at the end of the prosperous Ramesses reigns. These findings evidence thriving pre-Hellenistic settlements in Alexandria. During the Greek and Roman periods, we expound the largest Pb pollution ever encountered in ancient city sediments with Pb levels twice as high as those measured in contemporary industrialized estuaries. **Citation:** Véron, A., J. P. Goiran, C. Morhange, N. Marriner, and J. Y. Empereur (2006), Pollutant lead reveals the pre-Hellenistic occupation and ancient growth of Alexandria, Egypt, *Geophys. Res. Lett.*, *33*, L06409, doi:10.1029/2006GL025824.

[2] Alexandria, on the western Nile branch, is named after Alexander the Great who ordered its construction in 331 BC. In antiquity, the city flourished and its population is believed to have reached a few hundred thousand during the Roman apogee [*Empereur*, 1998; *Yoyotte et al.*, 1998]. The existence of a pre-Hellenistic settlement, named Rhakotis, has never been unequivocally reconciled and debate continues to rage in the literature [*Chauveau*, 1999; *Baines*, 2003]. Here we use Pb and its stable isotopes to geochemically infer: (1) the growth of ancient Alexandria and release of a pollutant into the harbor and (2) a significant Bronze Age geochemical imprint, inconsistent with the Alexandria ex nihilo hypothesis. Pb has proved to be a powerful tool in recognizing past Greek and Roman industrial activities [*Bintliff et al.*, 1992; *Pyatt et al.*, 2000; *Aberg et al.*, 2001] owing to its exclusive properties. Its corrosion resistance, malleability and low melting point mean it was extensively used in plumbing, architecture, weight and glass making, soldering, statue casting, ship building and fishing [*Lucas and Harris*, 1962; *Nriagu*, 1983]. Furthermore, its stable isotopes can be used to evidence anthropogenically dispersed pollutant Pb in sedimentary archives [*Chow et al.*, 1973; *Shirahata et al.*, 1980; *Ng and Patterson*, 1982; *Renberg et al.*, 1994; *Shotyk et al.*, 1998].

[3] Sediment cores were collected in Alexandria's eastern basin (core CV) and its tombolo (core CII). Detailed sedimentological descriptions are given by *Goiran* [2001]. Radiocarbon dates obtained from *Posidonia* fibers, shells fragments and *Cladocora* corals provide a working chronological framework. Dates were calibrated according to *Stuiver and Braziunas* [1993] and are presented in calendar years in Figures 1–2 and Table 1. Pb has four stable isotopes (mass = 204, 206, 207, 208) of which the last three are end-members of the natural uranium (U)-thorium (Th) decay chains. Pb ores display specific isotope ratios according to the age and the initial U-Th content of their primary geological reservoirs [*Doe*, 1970]. These ratios are preserved during smelting of the ores, distinguishing not only the geological origin of anthropogenically dispersed Pb but also pollutant versus crustal-derived Pb. Crustal sediments usually exhibit $^{206}Pb/^{207}Pb$ ratios higher than 1.2, whilst geochemical investigations on marine sediments demonstrate that small variations of 0.01 can correspond to changes in Pb sources and/or mineralogy [*Shirahata et al.*, 1980; *Ng and Patterson*, 1982]. For the Mediterranean, this tracing ability has been developed in archeology to differentiate the geographical provenance of copper (Cu), Silver (Ag) and Pb ores during antiquity [*Stos-Gale et al.*, 1996; *Yener et al.*, 1991; *Sayre et al.*, 1992]. Pb concentration and corresponding isotope ratios were measured in sediments from Alexandria's ancient harbor by thermal ionization mass spectrometry on a MAT Finnigan 262 at CEREGE. Mass fractionation was corrected using a SRM981 NIST standard. Pb concentrations were determined by isotope dilution using a ^{208}Pb isotope spike on separate aliquots. Uncertainties (including blanks and analytical errors) of Pb concentration and $^{206}Pb/^{207}Pb$ ratios were below 1% and 0.5‰ respectively. Crustal Enrichment Factors (EF) were calculated as the ratio of Pb to aluminum (Al, a major crustal component) in the sample against Pb/Al in crustal referenced material [*McLennan*, 1995]. EF below 3 are consistent with natural crust-derived Pb. Higher enrichment values invoke a different Pb source, namely of anthropogenic origin. Al was determined by ICP-AES Jobin Yvon at CEREGE.

[4] Core CV permits a high-resolution inquiry of anthropogenic impacts during antiquity. A low to moderate energy depositional environment is attested by decantation clays and *Posidonia* fibers [*Goiran et al.*, 2000]. Around 300 BC,

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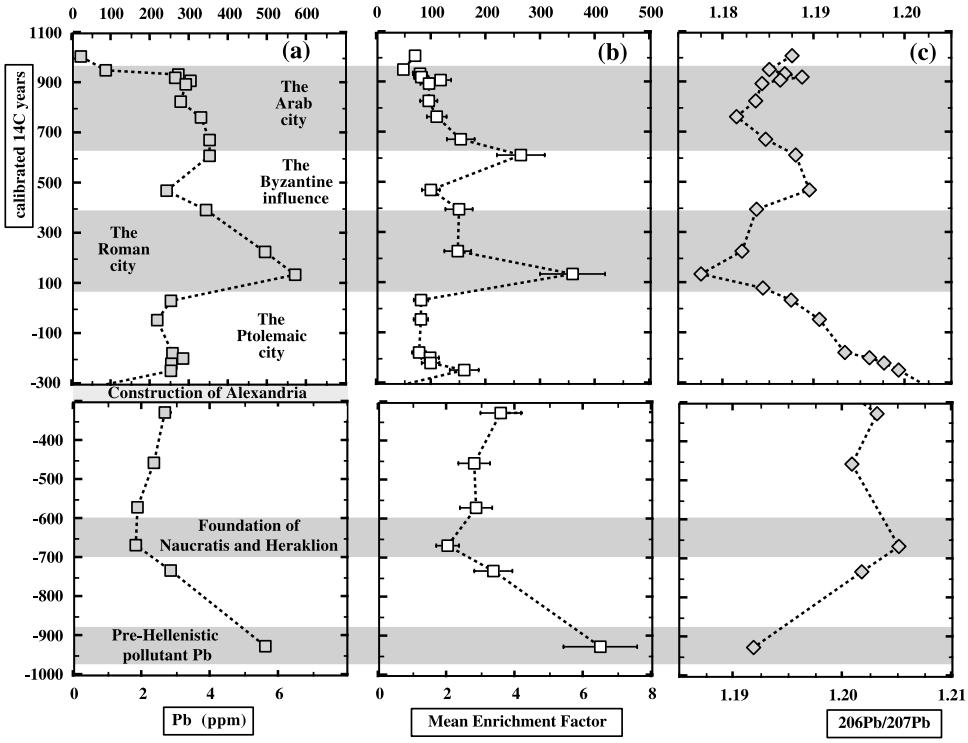

Figure 1. Changes in (a) Pb concentrations, (b) Pb enrichment factors and (c) isotope imprints in sediments from core CV.

we observe a clear change in Pb concentrations, shifting from 2–6 ppm to more than 200 ppm (Figure 1a). We link this unambiguous geochemical change to the Hellenistic foundation of the city. After this period, concentration levels of 200–300 ppm are recorded, rising rapidly to 600 ppm during the Roman apogee (100 AD to 300 AD). Calculated EF confirm these findings and closely follow Pb concentration patterns, with values superior to 50 after the city's foundation (Figure 1b). These concentrations are an order of magnitude higher than those measured in sediment deposits from Sidon, the only other ancient harbor to have undergone similar geochemical investigations [*Le Roux et al.*, 2003]. Greek and Roman Pb levels are equal to or higher than those today recorded in Alexandria's modern harbor (90 ± 20 ppm) [*Saad et al.*, 2003] and present day industrialized estuaries (50 to 200ppm) [*Shirahata et al.*, 1980; *Croudace and Cundy*, 1995; *Feng et al.*, 1998; *Li et al.*, 2000], reaching levels of toxicity similar to those defined for industrial soils. A clear isotopic contrast confirms the environmental impact of the city, with $^{206}Pb/^{207}Pb$ ratios shifting from crustal-derived values higher than 1.20, prior to 300 BC, to Mediterranean Pb-Cu-Ag ore signatures during the Greek (1.187–1.199, Greek ores from Laurion, Syphnos) and Roman (1.177–1.185, Turkish ores) periods (Figure 1c) [*Stos-Gale et al.*, 1996; *Yener et al.*, 1991; *Sayre et al.*, 1992; *Véron and Leroux*, 2004]. Isotope imprints from core CII are chronostratigraphically replicated by signatures from CV (Figure 2a). It is the first time that such high levels of environmental pollution have been recorded in an ancient marina, attesting the economic development of this metropolis with intensive commercial trade and metallurgic activities.

[5] Core CII records the accretion of the tombolo linking Pharos to the main city long before Alexander's arrival [*Goiran et al.*, 2000]. This result evidences the existence of a semi-enclosed and protected bay that could have been used as an anchorage haven. Pollen data (*Tamarix* and *Cerealia*) observed in this core indicate possible local human impact with a peak at the beginning of the 3rd millennium BC [*Goiran*, 2001]. Our Pb data sets allow three unequivocal geochemical phases to be defined for this core:

[6] - During the Predynastic period (~3800 BC to ~3000 BC), Pb concentrations and corresponding EF values are similar to those in pre-Hellenistic layers from core CV

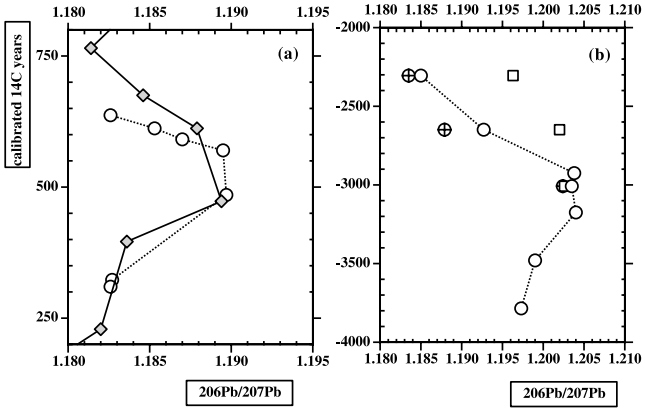

Figure 2. (a) comparison between Pb isotope imprints in core CV (closed lozenges) and core CII (open circles) in late Roman and Byzantine deposits; (b) Pb isotope imprints in core CII from pre-Hellenistic deposits for total Pb (open circles), acid leached (crossed circles) and residual (open squares) fractions.

Table 1. Geochemical Data Set for Sediment Deposits From Core CII (Alexandria Tombolo)[a]

Depth, m	Dates 14C Corr.	Pb, ppm	Al, %	EF	$^{206}Pb/^{207}Pb$	$^{208}Pb/^{206}Pb$
2.7	695 ± 77 AD	nd	nd	nd	nd	nd
3.2		27.9	nd	nd	1.1826	2.0863
3.9		38.6	2.14	6.6	1.1853	2.0836
4.2		52.4	0.25	78	1.1870	2.0811
4.5	570 ± 96 AD	24.8	0.24	38	1.1895	2.0773
4.8	485 ± 17 AD	31.5	0.23	50	1.1897	2.0805
5.1	323 ± 36 AD	147	0.69	78	1.1827	2.0857
5.7	310 ± 53AD	75.4	0.20	138	1.1826	2.0867
6.2	2307 ± 138 BC	8.66	0.60	5.3	1.1850	2.0858
6.6		3.33	1.22	1.0	1.1927	2.0755
6.9	2927 ± 88 BC	4.77	nd	nd	1.2038	2.0649
7.2		3.87	1.53	0.9	1.2035	2.0672
7.7		5.54	2.56	0.8	1.2040	2.0680
8.4		4.10	2.21	0.7	1.1990	2.0725
9.3	3783 ± 135 BC	4.16	1.37	1.1	1.1973	2.0711

[a]nd represents non determined data.

(Figures 1a and 1b and Table 1). These deposits show $^{206}Pb/^{207}Pb$ ratios close to 1.20, suggesting the absence of any pollutant Pb (Figure 2b).

[7] - For the Middle Bronze Age, ~2300 BC deposits display a significant Pb enrichment with EF of 5.3. Such enhancement is confirmed by an isotopic shift from 1.205 to 1.185 between 3000 BC and 2300 BC (Figure 2b). Acid leaching was performed on selected sediment samples to confirm the presence of anthropogenically-derived Pb. Owing to its unique isotopic imprint, a light leaching (HNO_3 0.5N) discriminates the occurrence of both residual crustal and leached pollutant Pb. Starting in ~2500 BC, sediments delineate significant differences between both fractions, with $^{206}Pb/^{207}Pb$ ratios close to 1.185 and higher than 1.195 for the leached and residual components respectively (Figure 2b). Middle Bronze Age release of leached Pb strongly suggests the onset of anthropogenic activities during the IVth dynasty. This period is characterized by a rise in the number of Nile delta settlements, as revealed by the development of durable trade between Egypt, Libya and the Sinai Peninsula [*Lucas and Harris*, 1962].

[8] During the Iron Age we observe a contamination peak as evidenced by EF of 6–7 and a $^{206}Pb/^{207}Pb$ ratio of 1.192 (Figure 1). Such geochemical evidence for pre-Hellenistic occupation of Alexandria is consistent with the prosperous Rameses reigns of the XIXth and XXth dynasties. At this time, the site lay within an administrative area protected by a series of fortresses constructed under Rameses II [*Brinton*, 1942; *Baines and Malek*, 2000]. It is posited that this settlement was named Raqote meaning "naval construction yard" [*Depauw*, 2000]. This revival at the end of the New Kingdom is followed by non contaminated overlying deposits. We postulate that the absence of any significant pollutant Pb between 800 BC and Alexander's arrival resulted from decline of the Egyptian empire during the Nubian, Assyrian and Persian invasions [*Butzer*, 1976].

[9] Previous findings allow the origin of this pollution to be reconciled (i.e., regional versus local). Geochemical investigations clearly demonstrate the large contribution of the Nile river to sediments accumulated in the Levantine basin [*Krom et al.*, 1999]. Meanwhile, exhaustive metal analyses of Holocene sediments deposited in the Nile delta does not evidence any noticeable Pb enrichment (as defined by crustal enrichment factors based on Al) [*Siegel et al.*, 1995]. Furthermore, the counter-clockwise Levantine gyre disperses Nile derived particles eastward, which prevent Nile material from reaching Alexandria bay. In addition, Alexandria bay sediments from core V do not record any immediate Pb enrichment following the foundation of close by trade cities (between 700 and 600BC), namely Naucratis, Menouthis and Heraklion [*Stanley et al.*, 2001]. These data all corroborate a local, in situ, provenance for pollutant Pb encountered in ancient Alexandrian sediments.

[10] Our data are not consistent with the foundation of Alexandria ex nihilo. Pre-Hellenistic Pb imprints suggest the existence of an in situ Bronze Age settlement as early as the IVth dynasty. It would indeed be paradoxical to assume that such a locally unique and geologically endowed site - an island breakwater sheltering a large marine bay on the extreme margin of the Nile delta - did not attract anterior societies. After an early ~1000 to 800 BC revival, relative decline of this settlement is recorded through to the 4th century BC. A pronounced rise in pollutant Pb concentrations around 330 BC concurs Alexander the Great's arrival, with extreme levels of pollution during the ensuing Hellenistic and Roman periods. These findings confirm pollutant Pb as an efficient tracer of ancient human activities and have widespread archeological implications regarding the early chapters of Alexandria's history.

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