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Hugo Delile[a,b,1], Janne Blichert-Toft[b,c], Jean-Philippe Goiran[d], Simon Keay[e], and Francis Albarède[b,c]

[a]Université Lumière Lyon 2, Centre National de la Recherche Scientifique-Unité Mixte de Recherche (CNRS UMR) 5600, 69676 Bron, France; [b]Ecole Normale Supérieure de Lyon, Université Claude Bernard Lyon 1, CNRS UMR 5276, 69007 Lyon, France; [c]Department of Earth Science, Rice University, Houston, TX 77005; [d]Maison de l'Orient et de la Méditerranée, CNRS UMR 5133, 69365 Lyon Cedex 7, France; and [e]Archeology, Faculty of Humanities, University of Southampton, Southampton SO17 1BF, Great Britain

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It is now universally accepted that utilization of lead for domestic purposes and water distribution presents a major health hazard. The ancient Roman world was unaware of these risks. How far the gigantic network of lead pipes used in ancient Rome compromised public health in the city is unknown. Lead isotopes in sediments from the harbor of Imperial Rome register the presence of a strong anthropogenic component during the beginning of the Common Era and the Early Middle Ages. They demonstrate that the lead pipes of the water distribution system increased Pb contents in drinking water of the capital city by up to two orders of magnitude over the natural background. The Pb isotope record shows that the discontinuities in the pollution of the Tiber by lead are intimately entwined with the major issues affecting Late Antique Rome and its water distribution system.

harbor geoarcheology | paleopollution | Late Holocene | ore provenance | sedimentology

Statistics on demography, money supply and metal circulation, life and health standards, and many other social parameters required to understand modern history are largely missing from the written record of the ancient past. For example, the apparently simple question of how the population of ancient Rome evolved is still unresolved (1, 2), prompting the design of indirect estimates (3). Another well-publicized problem illustrating the lack of primary sources of accurate information is the decade-old debate on Pb poisoning of the high society of Rome, either by lead water pipes or grape juice concoctions prepared in lead cups (4–9). Here we focus on the condition of Pb in the public waters of ancient Rome. Lead is regarded as a powerful and ubiquitous indicator of the manufacturing status of a society. For example, a surge in Pb concentrations in the Greenland ice-core record was correlated with the height of the Roman Empire (10). Three out of the four existing Pb isotopes are rapidly modified by the radioactive decay of natural uranium or thorium over geological time. The mining of ores from geologically diverse areas produces metallic Pb with variable isotopic abundances that depend on the tectonic age and the Th/U and U/Pb ratios of the mining district. Archeologists interested in the provenance of artifacts routinely tap this wealth of information (11). To explore how the supply of metals from all over the Roman world and their utilization may have affected the nearby environment of ancient Rome, the present work sets out to investigate the isotope compositions of Pb in sediment cores from the Trajanic harbor basin at Portus, the maritime port of Imperial Rome, and the channel connecting Portus with the Tiber (Canale Romano) (Fig. S1). Harbors are excellent sedimentary traps. The record of human Pb pollution from the time that the harbor basin was excavated (*ca.* 112 AD) and well into the Middle Ages offers a new historical, *ca.* 1,000 y-long perspective on the evolution of Pb released by Rome, its water distribution system, and the major disruptive events that affected the life of the capital city and its harbor.

In 42 AD Claudius started the construction of an open coastal port to compensate for the long-standing shortcomings of the existing system for supplying Rome from the Mediterranean, notably the small scale of the harbor and anchorage facilities at Ostia and the long route of communication with the principal maritime port at Puteoli (Pozzuoli) on the Bay of Naples (12). The inland ~0.4 km^2 Trajanic basin, which was excavated in the early years of the second century AD in response to the growing demands of an expanding population in Rome, offered both safe mooring to sea-going merchant ships and immense warehouses and other buildings (13–15). Communication between the Claudian and Trajanic basins was facilitated by an entrance channel, into which the *ca.* 9-m-long core TR14 was drilled. Up until the Middle Ages, the Trajanic basin was also accessed from a man-made branch of the Tiber (Fossa Traiana; what is now the Fiumicino Canal) by means of the Canale Traverso. The transport of sand and silt sediments from this channel to the Trajanic Harbor has been attested to by sedimentological, geochemical, and ostracod analyses (16–18). The now filled-in Canale Romano, which ran past the southwestern side of the Trajanic basin toward the Tiber, was used to carry cargoes transshipped on to river-going craft bound for Rome (15). A 13-m-long core labeled CN1 was drilled into the sediments of the Canale Romano. The detailed sedimentology and geochemistry of core TR14 are given elsewhere together with ^{14}C ages (18). Some ^{14}C dates likewise were obtained for core CN1 (Table S1). For reference and modeling purposes, the bedload of the modern Tiber between Rome and the Tiber delta was also sampled (Table S2), as were five different Roman Pb water pipes (*fistulæ*) collected in Rome and dating to between the first and the second centuries AD (Fig. S2). In all, 42 samples from TR14, 37 samples from CN1, 6 samples from the Tiber bedload, and 10 samples from the five Roman *fistulæ* were measured for their Pb isotope compositions at the Ecole Normale Supérieure de Lyon.

Significance

Thirty years ago, Jerome Nriagu argued in a milestone paper that Roman civilization collapsed as a result of lead poisoning. Clair Patterson, the scientist who convinced governments to ban lead from gasoline, enthusiastically endorsed this idea, which nevertheless triggered a volley of publications aimed at refuting it. Although today lead is no longer seen as the prime culprit of Rome's demise, its status in the system of water distribution by lead pipes (*fistulæ*) still stands as a major public health issue. By measuring Pb isotope compositions of sediments from the Tiber River and the Trajanic Harbor, the present work shows that "tap water" from ancient Rome had 100 times more lead than local spring waters.

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[1]To whom correspondence should be addressed. E-mail: hdelile@gmail.com.

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Results and Discussion

The TR14 core can be broken down into successive sedimentary units corresponding to different time slices: (*i*) preharbor up to *ca.* 100 AD, (*ii*) Early Empire up to *ca.* 250 AD, (*iii*) Late Empire up to *ca.* 500 AD, (*iv*) Early Middle Ages up to *ca.* 800 AD, and (*v*) Late Middle Ages (see the "Historical period age-depth model" columns in Fig. 1). Age boundaries between units may be uncertain by up to 100 y. Silts and sands dominate sediment mineralogy. The preharbor sequence attests to deposition in an environment of deltaic progradation (19). The construction of the harbor brings about a sharp sedimentological change and marks the beginning of the harbor mud deposits. A well-stratified ~50-cm-thick layer within the Early Empire deposits (753 cm) displaying well-preserved shells does not appear in other cores and may signal local dredging (18, 20). The layer corresponding to the Early Middle Ages contains more carbonates and ostracods of brackish affinity than the rest of the core. At the top, the sediments from the Late Middle Ages horizon are characteristic of flood plain deposits (17, 18).

In Fig. 1, two different representations of lead isotope variations in TR14 and CN1 have been used: First (Fig. 1*A*), the conventional raw isotopic ratios in which ^{206}Pb is kept as the denominator; and, second (Fig. 1*B*), a derived set of geologically informed parameters which will now be explained. In compliance

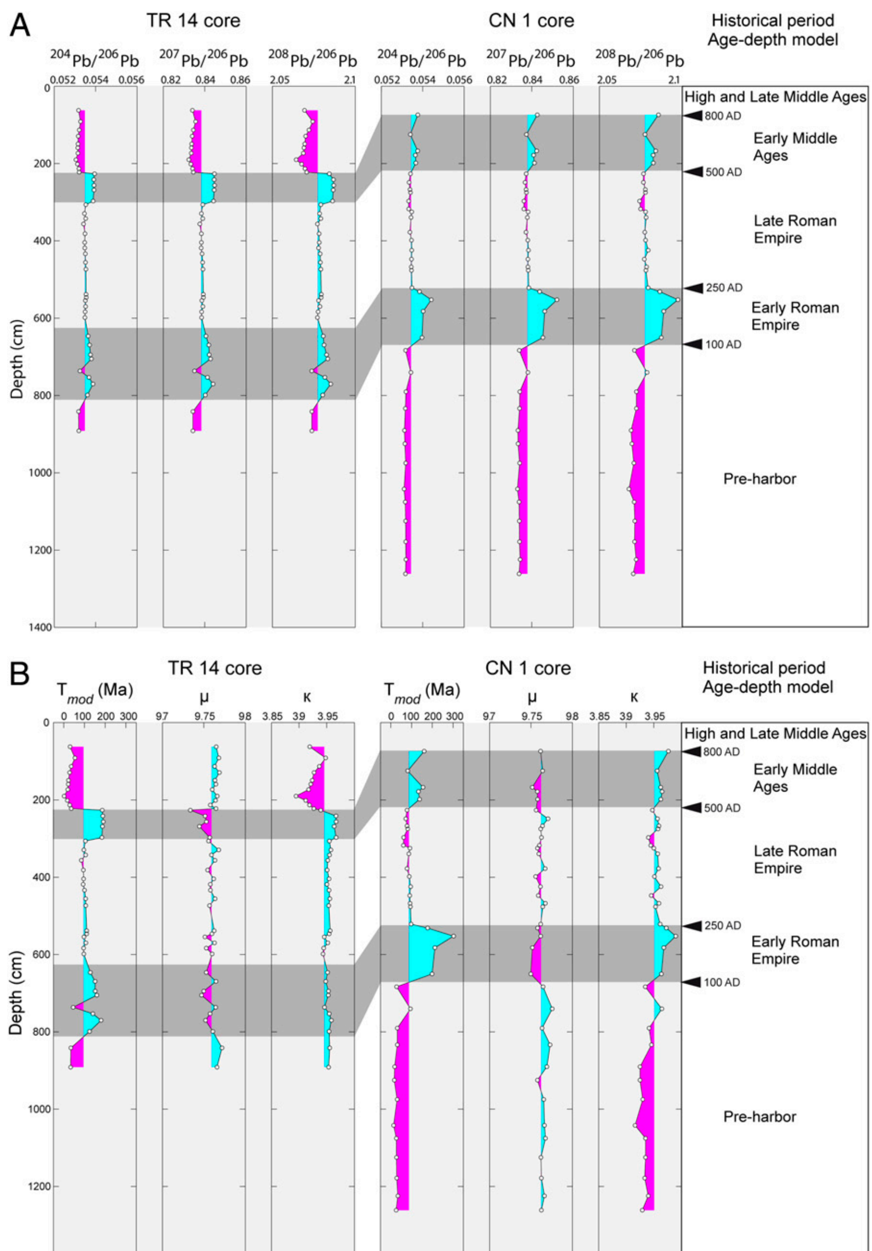

Fig. 1. Chronostratigraphic evolution of (*A*) the raw isotopic ratios of the cores TR14 and CN1 and (*B*) the geological parameters T_{mod}, $^{238}U/^{204}Pb$ (μ), and $^{232}Th/^{238}U$ (κ) derived from the raw isotope ratios as described in the main text. The different time slice boundaries (indicated with black arrowheads and highlighted by alternating light and dark gray shading for better visibility) derived from the age-depth model of core TR14 (18) coincide with major Pb isotope compositional shifts prominent in both cores. Hercynian Pb shows up during the Early Roman Empire and Early Middle Ages. In contrast, low-Th/U natural Pb dominates both the preharbor sequence and the flood plain deposits at the top of TR14.

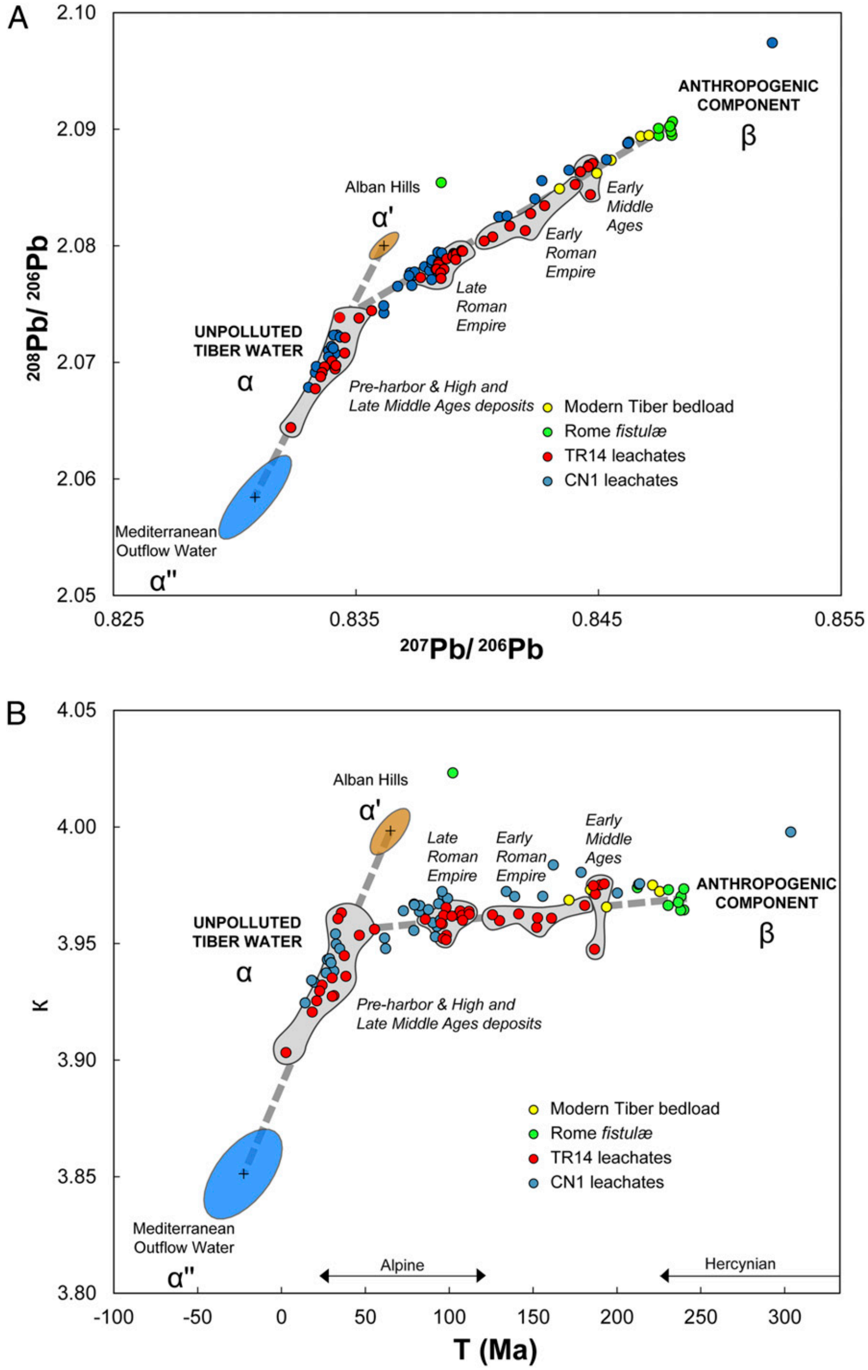

Fig. 2. (*A*) Lead isotope ratios ($^{207}Pb/^{206}Pb$ vs. $^{208}Pb/^{206}Pb$) and (*B*) geological parameters (κ vs. T_{mod}) for the leached samples from cores TR14 (red) and CN1 (blue), the modern Tiber bedload (yellow), and Rome *fistulæ* (green). The gray fields correspond to the light and dark gray shaded time slice bands of Fig. 1 and overlap the samples from core TR14 in accordance with the respective historical periods. The two mixing lines (gray dashes) connect, respectively, α and β on the one hand, and α′ and α″ on the other. The α end-member corresponds to unpolluted Tiber water and is composed of the Mediterranean outflow water (α″, blue ellipse) (30) and volcanic rocks from the Alban Hills (α′, orange ellipse) (28, 29). β is the anthropogenic end-member.

with literature (e.g., ref. 21), we searched Pb isotope databases for potential sources of ores matching the Pb isotope compositions of archeological artifacts and sediment samples (Fig. S3). In addition, the geological province to which a particular Pb sample belongs can often be inferred from a conversion of its isotope compositions into a set of geologically informed parameters, the Pb model age T_{mod}, and the apparent $^{238}U/^{204}Pb$ (μ) and $^{232}Th/^{238}U$ (κ) ratios (e.g., refs. 22–24). T_{mod} reflects the tectonic age of the crustal segments in which ore deposits occur. In Europe, crustal segments of Alpine ages (30–120 Ma) contrast with Hercynian (240 Ma and older) and early Paleozoic (>450 Ma) segments. μ and κ are parameters that tend to increase with crustal depth. Typically, κ is higher in crustal segments that lost their shallow levels by erosion or tectonic denudation, such as in Iberia, southern France, and the eastern Alps. Fig. S4 shows that these parameters can be used to divide Europe into coherent regions, which justifies using T_{mod}, μ, and κ for provenance purposes (24–26). T_{mod}, μ, and κ in turn provide a rapid characterization of the geological environment in which the ores formed. Ores formed by remobilization of metal from the underlying basement and hosted in sediments, such as Mississippi Valley type deposits, may to some extent challenge a simple interpretation of model ages. Fig. S4 shows, however, that, overall, the connection between Pb model ages and the tectonic age of the local crystalline basement remains very strong. The broad relationship between T_{mod}, μ, and κ tectonic provinces is compelling and holds particularly true for southern Europe (27).

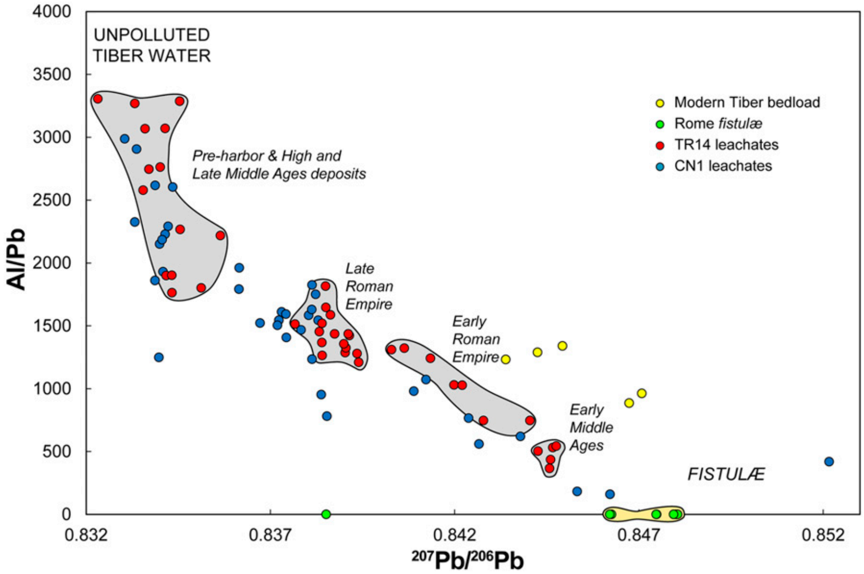

Fig. 3. Al/Pb vs. $^{207}Pb/^{206}Pb$ showing the TR14 (red) and CN1 (blue) core leachates, the modern Tiber bedload (yellow), and the Rome *fistulæ* (green). The beige field corresponds to the anthropogenic component characterized by the *fistulæ*. Symbols and parameters are as in Fig. 2. This plot shows that the Al-free, and therefore suspension-free, water component has the same Pb isotope composition as the *fistulæ* and therefore corresponds to clear water from the city water distribution system.

Principal component analysis of the 3D Pb isotopic data shows that >99% of the variance is accounted for by two principal components and, therefore, that the data plot in a plane spanning any 3D space of Pb isotopic ratios. Cores TR14 and CN1 define indistinguishable planes, which allows the Pb isotope data to be merged into a single dataset. As illustrated by the $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$ plot of Fig. 2*A*, the isotope composition of Pb in leachates form two coplanar alignments, which are most straightforwardly accounted for by the mixing of components of different origins.

In Fig. 2 *A* and *B*, the component labeled α located at the intersection (the kink) of the two trends is ubiquitous in both cores including the preharbor and Late Middle Ages deposits. It is also an end-member in plots of Pb isotope ratios from leaching residues (Fig. S5 *A* and *B*). This component therefore reflects Pb naturally present in Tiber water. It can itself be broken down into a mixture of two local low-$^{207}Pb/^{206}Pb$ sources, the component α′ originating from the recent volcanic rocks of the Alban Hills (28, 29), and the component α″, which is very similar to Pb dissolved in modern Mediterranean seawater (30) and released by erosion of recent limestones from the Apennines.

The anthropogenic nature of the third component β becomes apparent when plotting Al/Pb (data from ref. 18) as a function of $^{207}Pb/^{206}Pb$ (Fig. 3). The α–β alignment intersects the *x* axis at the value of $^{207}Pb/^{206}Pb$ of the *fistulæ* (Al/Pb ≈0), which shows that the contaminant is essentially pure lead from Al-free and therefore suspension-free water. As with raw isotopic ratios, a plot of κ vs. T_{mod} (Fig. 2*B*) shows a bundle of alignments consistent with the observations from isotopic ratios. The alignment trending toward high $^{207}Pb/^{206}Pb$ and old model ages reveals that Pb component β is of Hercynian (or Variscan; T_{mod} ≈250 Ma) affinity with rather high κ values. Hercynian Pb is absent from peninsular Italy, and the Apennines formed less than 20 Ma ago (31) from recent sediments and volcanic rocks. The Pb component β therefore, clearly being foreign to peninsular Italy, should rather be traced to southwestern Spain, the Massif Central of France, the eastern Alps, Eifel in Germany, the Pennines in England, and Macedonia (Fig. S3). Among these potential sources, only some of them are consistent with the known maritime freight routes, which are punctuated by frequent shipwrecks loaded with Pb ingots (32–34), and with the known period and output of mine exploitation during the Late Republican Period and the Early Roman Empire (e.g., refs. 21 and 35). An unexpected observation is the lack of signal from the productive and geologically young mining areas of the Spanish Betics (Carthagene). It is most likely that the Pb used for water management in Rome had been mined in the Spanish Sierra Morena, the English Pennines, the German Eifel, or the French Massif Central.

The isotope composition of component β is remarkably consistent with the data on four of the five lead *fistulæ* analyzed in this work. Component β is still conspicuous in the leachates from the modern Tiber bottom sediments, which suggests that to this day old Pb pollution still permeates the bedload sediment. The anthropogenic origin of the Hercynian Pb component β is also attested to by the comparison of leachates and residues: $^{207}Pb/^{206}Pb$ is, with the sole exception of the deepest sample, higher in leachates than in residues (Fig. 4). Leaching therefore releases older labile Pb from a solid residue of much younger geological age.

Lead pollution of the Tiber River can be evaluated in a simple way by using

$$\varphi_{fist} = \frac{(^{207}\mathrm{Pb}/^{206}\mathrm{Pb})_{riv} - (^{207}\mathrm{Pb}/^{206}\mathrm{Pb})_{nat}}{(^{207}\mathrm{Pb}/^{206}\mathrm{Pb})_{fist} - (^{207}\mathrm{Pb}/^{206}\mathrm{Pb})_{nat}},$$

where φ_{fist} is the fraction of Pb in river water derived from Pb *fistulæ*. It has been estimated that the proportion f_{fist} of Tiber water running through the aqueducts was about 3% at the peak of the Roman Empire (36). It can therefore be deduced that *fistulæ* increased Pb in the water distributed in Rome over the natural level by a factor of about 40, 14, and 105 for the Early Empire, Late Empire, and High Middle Ages, respectively (*SI Materials and Methods*). Although the value of f_{fist} is only an educated guess pertinent to a given period, using different numbers does not significantly affect the relative levels of Pb pollution deduced. These levels are maximum values because they characterize the final output of the water system to the Tiber while most Roman citizens would have used drinking water that was tapped, whether legally or illegally, all along the water distribution system (36). The inferred increases of Pb in the water of the Roman distribution system unquestionably attest to general lead pollution of Roman drinking water but the Pb concentrations at issue are unlikely to have represented a major health risk (9).

Evidence bearing on the timeline of anthropogenic pollution in the Rome area can be derived from the sequence of Pb isotope characteristics (Fig. 2). Lead in preharbor sediments is of

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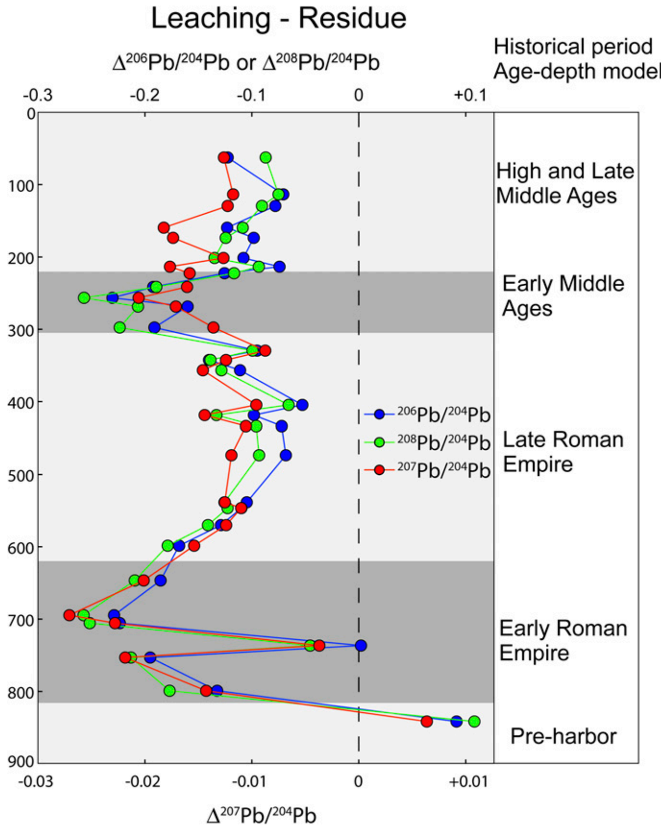

Fig. 4. TR14 downcore behavior of the isotopic contrast (ΔPb isotope ratios) between the residue and the leachate. Increasing distance from the zero dashed line (left side of the figure) indicates increasingly predominant anthropogenic impact, which is strongest during the Early Roman Empire and the Early Middle Ages. Time slice model as in Fig. 1.

natural origin (trend $\alpha'-\alpha''$). Excavation of basin deposits dating to the period of the Early Roman Empire coincides with both a surge of Hercynian Pb in leachates and a dramatic drop in the isotopic contrast between the residue and the leachate (trend $\alpha-\beta$) (Fig. 4). At this time, the Roman Empire reached the height of its conquests, especially in western European territories such as Britain (Fig. S3). The isotopic contrast between the fractions rapidly diminishes, although quite smoothly, from the Early to the Late Roman Imperial periods. This change is largely accounted for by the dramatically smaller contribution of anthropogenic Pb to leachates and therefore by a lesser pollution of Tiber water. One interpretation of this may be a redirection of spring water away from the lead pipes of Rome, in some way related to the controversial decline of the population (3, 37) or to a poorly documented deterioration of the water distribution system.

At the end of the Late Roman Empire and throughout the Early Middle Ages section of TR14, the isotopic difference between the leachate and the residue bounces back and the presence of a rather homogenous anthropogenic component rich in Hercynian Pb again becomes prevalent in leachates (Figs. 2 and 4). The discontinuity appears in both the TR14 and CN1 cores (Fig. 1). The end of the Early Middle Ages section (~800 AD) is also brutal and signals the return of uncontaminated Pb in the Tiber (trend $\alpha'-\alpha''$). The persistence of Hercynian Pb in the bedload of the modern Tiber nevertheless indicates that centuries of contamination, possibly in the form of Pb carbonates, left a lasting imprint on the river sediments.

The consistency of the Pb isotope results from the CN1 core, which is expected to carry a straightforward Tiber signal, with those from the TR14 core is rather good despite the latter being susceptible to both harbor activity and input of water from the Portus aqueduct, which has its source in the vicinity of modern Ponte Galeria. Both cores reflect the presence of a Hercynian end-member and coincide on the timing of major isotopic shifts. κ values in the CN1 core may, however, be marginally higher than those in TR14, especially during the Early Empire. It is unfortunate that the Pb isotope database on pipes used for water distribution is still too limited to identify such small differences with confidence.

The extensive nature of the harbor installations calls for additional work beyond the 95 samples of sediment core, bedload, and Pb pipes from Portus and the Tiber analyzed in this study to demonstrate unambiguously that the observed discontinuities in the Pb isotope and overall geochemical record correspond to catastrophic disruptions of Portus activity. Although the coastal position of the port leaves the Trajanic basin vulnerable to river vagaries and maritime hazards, the lack of coarse gravels and sediment sorting, combined with the good preservation of the delicate ostracod shells, are strong evidence against exceptional floods, storms, and tsunamis. The age-depth model (18) is certainly evocative of some critical dates of Roman history. As speculated by Delile et al. (18) based on the ^{14}C record of TR14 and adjacent cores, transitions between units may be correlated with the initial excavation of the Trajanic basin (by *ca.* 112 AD), the continued use of the port during the third century (*ca.* 250 AD), the gradual fortification and contraction of the port in the later fifth and earlier sixth centuries (*ca.* 500 AD), and the transition to the post-Byzantine period. The later fifth and sixth century transition is coeval with Belisarius' fixing of the decommissioned aqueducts of Rome (38) at the end of the Gothic Wars (535–554 AD). Byzantine repairs of the water distribution system may have remobilized massive amounts of corrosion products from abandoned lead pipes in which water may have stagnated for protracted lengths of time. Although a causal relationship cannot be formally demonstrated, the discontinuities in the cores at Portus seem contemporaneous with historically documented events such as the struggle for the control of the port between Gothic and Byzantine forces (536–552 AD) and the damages inflicted to the water distribution system during the Arab sack of Rome in the mid-ninth century. Further work is needed to learn whether the causes of Portus' demise were natural, with the harbor finally falling into disuse on account of flood plain deposits, possibly after the major floods of 856 AD (39–41), or a consequence of military events (39).

Conclusions

This work has shown that the labile fraction of sediments from Portus and the Tiber bedload attests to pervasive Pb contamination of river water by the Pb plumbing controlling water distribution in Rome. Lead pollution of "tap water" in Roman times is clearly measurable, but unlikely to have been truly harmful. The discontinuities punctuating the Pb isotope record provide a strong background against which ideas about the changing character of the port can be tested.

Materials and Methods

After removal of the coarse gravel fraction, 500 mg of sample were crushed and treated with chloroform to remove most of the abundant organic fraction. The residue was rinsed and leached in dilute HBr. Because lead pipe corrosion products, such as Pb carbonates (42, 43), were suspected to be present in the sediments and carry a signal from aqueducts, no attempt was made at using the more specific protocols developed to selectively extract hydroxide coatings (e.g., ref. 44). Lead from the leachates was purified on an ion exchange resin using HBr as eluent of the sample matrix and HCl to elute Pb. The amounts of Pb extracted were large (>1 μg) and orders of magnitude above the blank of the procedure (~20 pg). Lead isotope compositions were analyzed by multicollector inductively-coupled plasma mass spectrometry on both the residues and the leachates of the samples from TR14 and the results were so systematic that no further attempt was made to also measure the residues from CN1 (Table S2).

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