

A lead isotope perspective on urban development in ancient Naples

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The influence of a sophisticated water distribution system on urban development in Roman times is tested against the impact of Vesuvius volcanic activity, in particular the great eruption of AD 79, on all of the ancient cities of the Bay of Naples (Neapolis). Written accounts on urbanization outside of Rome are scarce and the archaeological record sketchy, especially during the tumultuous fifth and sixth centuries AD when Neapolis became the dominant city in the region. Here we show that isotopic ratios of lead measured on a well-dated sedimentary sequence from Neapolis' harbor covering the first six centuries CE have recorded how the AD 79 eruption was followed by a complete overhaul of Neapolis' water supply network. The Pb isotopic signatures of the sediments further reveal that the previously steady growth of Neapolis' water distribution system ceased during the collapse of the fifth century AD, although vital repairs to this critical infrastructure were still carried out in the aftermath of invasions and volcanic eruptions.

AD 79 Somma-Vesuvius eruption | Pb isotopes | harbor geoarchaeology | Neapolis | paleo-pollution

Urban centers have always been critically dependent on a stable water supply, and ancient cities relying on masonry aqueducts were particularly vulnerable to the disruption of their water distribution system by earthquakes and volcanic eruptions (1). The archaeological record of the major eruption of Vesuvius in AD 79 and its effect on the water supply of Naples, then known as Neapolis, and its neighboring cities illustrates well how efficiently the Roman world was able to mitigate the effects of major disasters on the daily life of its population.

Neapolis: Water Supply and Volcanism

Neapolis and the surrounding region were supplied with water from the Aqua Augusta or Serino aqueduct, built during the reign of Augustus between 27 BC and AD 10 (2, 3). The Augusta was a regional network supplying eight or nine cities, as well as numerous villas, through multiple branches (Fig. 1A): Nola, possibly Pompeii, Acerrae, Atella, Neapolis, Puteoli, Cumae, Baiiae, and Misenum (2, 4). The total length of the aqueduct, including its branches, was ~140 km. The construction of this monumental hydraulic network helped meet a need to secure the water supply for the strategic region of Campania during a critical period: the establishment of the Principate (2). The aim of the Augusta was to provide water to naval harbors (first Portus Iulius and later Misenum) and the commercial harbor of Puteoli, one of the busiest centers of trade in the Roman Empire (5), as well as to cities, *coloniae*, and villas of influential individuals. At an unknown time between the fifth century BC and the Middle Ages, the Bolla aqueduct (Fig. 1A) was constructed to bring additional water to Neapolis (3).

One of the challenges in maintaining the Augusta and, with it, the integrity of the water supply of the heavily settled area around Neapolis, was counteracting the slow movements of the ground associated with the activity of volcanic systems, known as

bradyseism. Roman water distribution systems consisted of large stone or concrete aqueducts, whose water was, in the western half of the empire at least, distributed to fountains and baths, residences, and other buildings by a large network of *fistulae*, lead pipes of different diameters but typically centimeter-sized. The availability of piped water at Pompeii, and more broadly at all of the cities of the Bay of Naples supplied by the Aqua Augusta, in response to the impacts of the AD 79 volcanic eruption of Somma-Vesuvius, is a matter of debate (6, 7). Interpretations of archaeological evidence from Pompeii itself disagree as to whether the town was receiving any piped water shortly before the eruption, whereas other viewpoints have emphasized the damaging effect of changes in topography preceding the AD 79 eruption on the aqueduct supplying Pompeii (6, 8). Repairs to the aqueduct channels at Ponte Tirone, near where the Pompeii aqueduct may have connected with the Aqua Augusta supplying Naples, have been interpreted as a remediation of the effects of both pre- and post-AD 79 bradyseism on this aqueduct's performance (3, 6).

Lead Contamination in the Harbor of Neapolis

To investigate the potential disruption of water supply around the Bay of Naples in the wake of the AD 79 eruption, we measured Pb isotopic compositions and elemental concentrations of the harbor sediments of Neapolis (Fig. 1B and C). Stratigraphic sections were made available as part of the archaeological excavation of the ancient harbor of Naples undertaken at Piazza Municipio by

Significance

A well-dated sedimentary sequence from the ancient harbor of Naples sheds new light on an old problem: could the great AD 79 Vesuvius eruption have affected the water supply of the cities around the Bay of Naples? We here show, using Pb isotopes, that this volcanic catastrophe not only destroyed the urban lead pipe water supply network, but that it took the Roman administration several decades to replace it, and that the commissioning of the new system, once built, occurred nearly instantaneously. Moreover, discontinuities in the Pb isotopic record of the harbor deposits prove a powerful tool for tracking both Naples' urbanization and later major conflicts at the end of the Roman period and in early Byzantine times.

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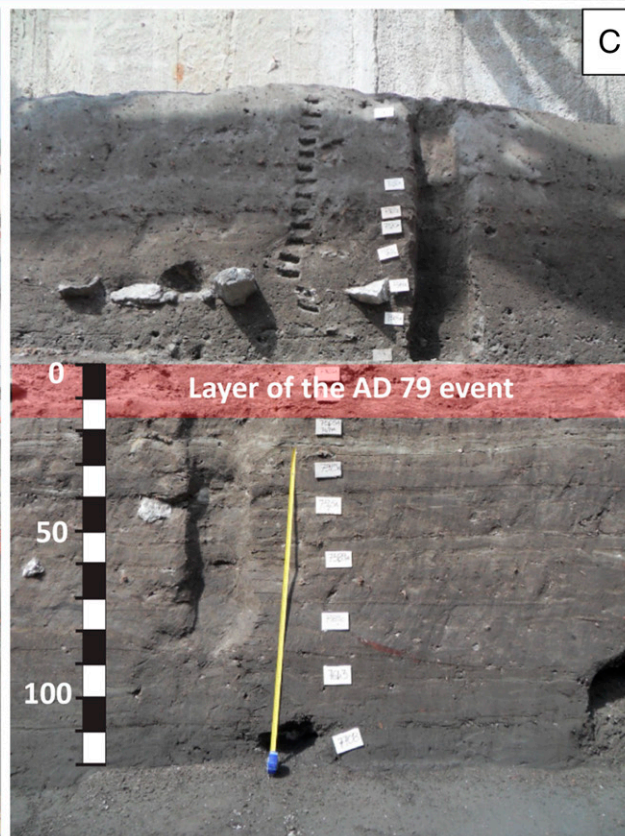
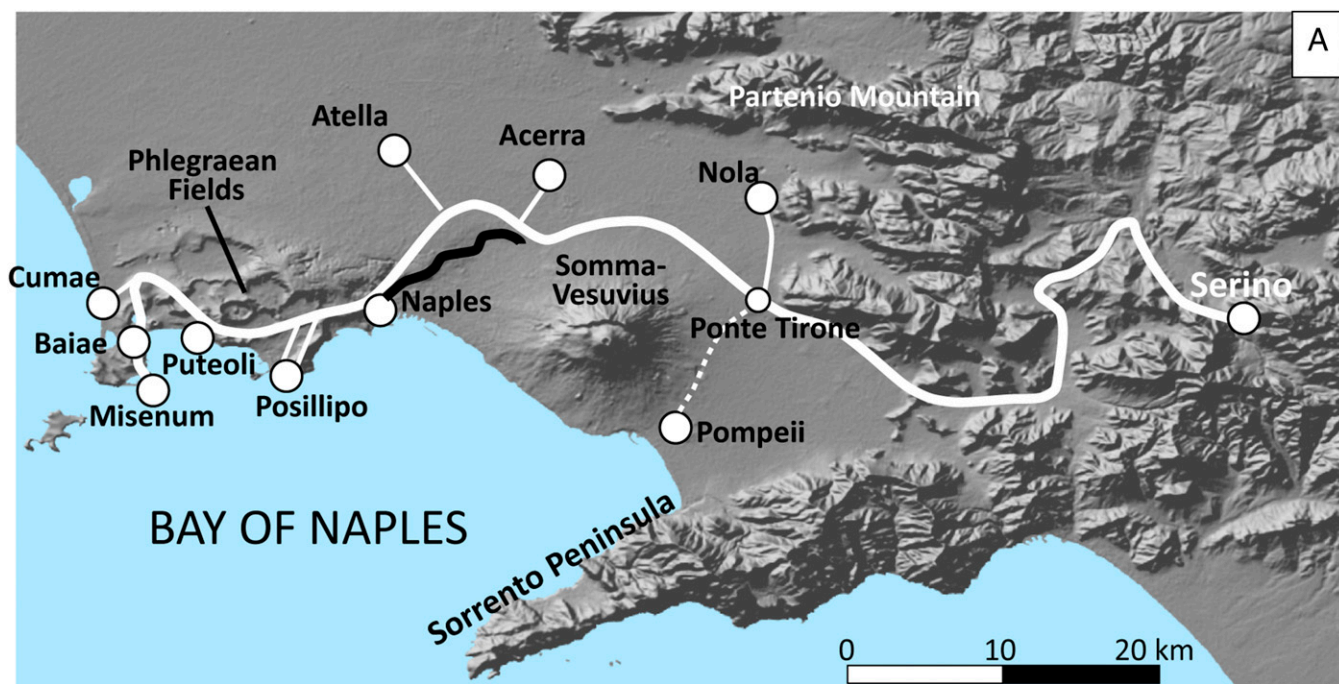


Fig. 1. Location of the study area. Neapolis is located halfway between two volcanic areas, Somma-Vesuvius and the Phlegraean Fields (A). The white bold line in A shows the main route of the Aqua Augusta aqueduct with its branches represented by the thinner white lines. The dotted line indicates the uncertainty over whether Pompeii was supplied by the Aqua Augusta. The black line shows the main route of the Bolla aqueduct. The archaeological excavation of the ancient harbor of Naples is located a few meters below current sea level in front of Piazza Municipio (B). Seen on the left side of the photo is the harbor dock composed of two levels: a lower level dating back to the Hellenistic period and an upper level raised in the Augustan period because of a rise in sea level. (C) An example of the harbor stratigraphic section investigated in this study and located in the eastern part of the excavation site.

the “Soprintendenza Speciale per i Beni Archeologici di Napoli e Pompei” (9). Ongoing excavation since 2011 allowed us to sample a 5.5-m-long sediment sequence (Fig. 1C). These deposits

are well dated by archaeological materials (9–13), with better precision than ^{14}C or optically stimulated luminescence dating, and they record the history of the city during the first six centuries

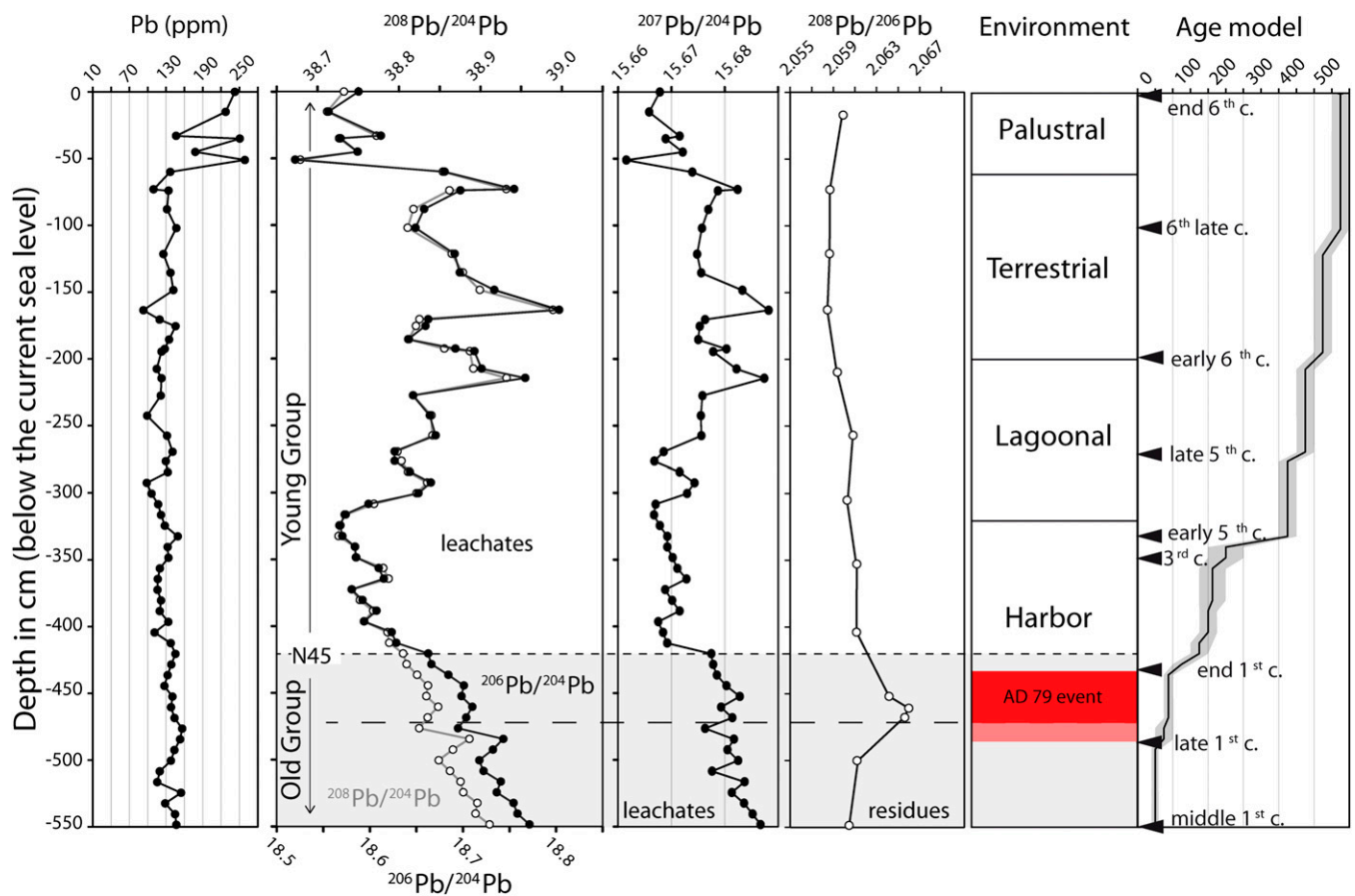


Fig. 2. Downcore variations of $^{208}\text{Pb}/^{204}\text{Pb}$, $^{206}\text{Pb}/^{204}\text{Pb}$, and $^{207}\text{Pb}/^{204}\text{Pb}$ in leachates, $^{208}\text{Pb}/^{206}\text{Pb}$ in residues, and Pb concentrations. The Young Group consists of all of the samples above layer N45, and the Old Group of all of the samples below. The different paleo-environmental units are also indicated (9, 12, 13). The dates supporting the Age Model of the section were provided by archaeological materials (9–13). The tephra unit of the AD 79 event is indicated by red shading. This latter is identified both geochemically (dark-red shading), by the cluster of the three samples constituting the upper end-member of the unpolluted water mixing line (component α ; see Fig. 3); sedimentologically (light-red shading), by specific sedimentological features (pumice stones); and archaeologically, by consistent archaeological dates (i.e., the second and third date from the bottom of the section). The parallel drift of $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ through time toward geologically old Pb reflects an increasing influence of pollution by the Pb pipe network (*fistulae*) and is a measure of urban development.

CE (Fig. 2). The level corresponding to the AD 79 eruption is located between -485 and -436 cm. The sediment at that level is heterogeneous and easily recognizable by shell debris, abundant fragments of wood, *Posidonia*, and pottery, as well as large numbers of rolled pumice pebbles (Plinian pumice lapilli fallout) (14).

Lead concentrations in the Neapolis harbor sediments (93–259 ppm) and the enrichment factor (EF_{Pb}) (Table S1) are similar to previous observations of contaminated sediments (15–17), amounting to excesses of Pb relative to natural Pb concentration levels by a factor of 3–5, deemed to signal anthropogenic pollution (15). The lack of significant variations in Pb abundances throughout the core, with the exception of the top 50 cm, shows that uncontaminated preharbor layers have not been found. The lack of a preharbor unit has been attributed to the dredging of the bottom sediments during the late fourth century/middle third century BC (9, 11, 12, 18), which is attested to by scars in the underlying Yellow Tuff bedrock.

Lead isotope compositions were measured on the sediments to separate the local environmental Pb background residing in minerals from the labile imported components. Samples were leached in chloroform and dilute HBr, and Pb isotope ratios measured on the leachates and their residues. The AD 79 layers stand out as a spike in $^{208}\text{Pb}/^{206}\text{Pb}$ in the residues at -461 ,

and -453 cm (N49R, N50R, and N51R) and in the leachates as a small dip in $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ (Fig. 2). In the very illustrative plot of $^{208}\text{Pb}/^{206}\text{Pb}$ vs. $^{204}\text{Pb}/^{206}\text{Pb}$ (Fig. 3A), the residues form an alignment distinct from the other two alignments defined by the leachates. The three samples of Neapolitan Yellow Tuff substratum of the harbor fall at the lower end of the residue field (Fig. 3A), hereafter referred to as component α (Fig. 3A).

The leachates form two parallel mixing arrays corresponding to two identifiable sets of samples: the “Old Group,” which includes all of the lowermost layers up to sample N45 (-421 cm), and the “Young Group,” which comprises all of the samples above N45 (Fig. 2). The calculated intersections of both leachate arrays with the residue array (star symbols in Fig. 3) suggest that the leachate and the residue contain a common component, probably from a readily leachable mineral phase present in the local watershed, typically carbonate.

We converted the Pb isotope compositions into their Pb model age T_{mod} and the time-integrated $^{238}\text{U}/^{204}\text{Pb}$ (μ) and $^{232}\text{Th}/^{238}\text{U}$ (κ) ratios (Table S1), using the equations of Albarède et al. (19). The unique information carried by these alternative coordinates relative to those of raw Pb isotope ratios has been demonstrated in several previous studies (see refs. 19–22). Lead model ages T_{mod} (in million years, Ma) are proxies for the tectonic age of the

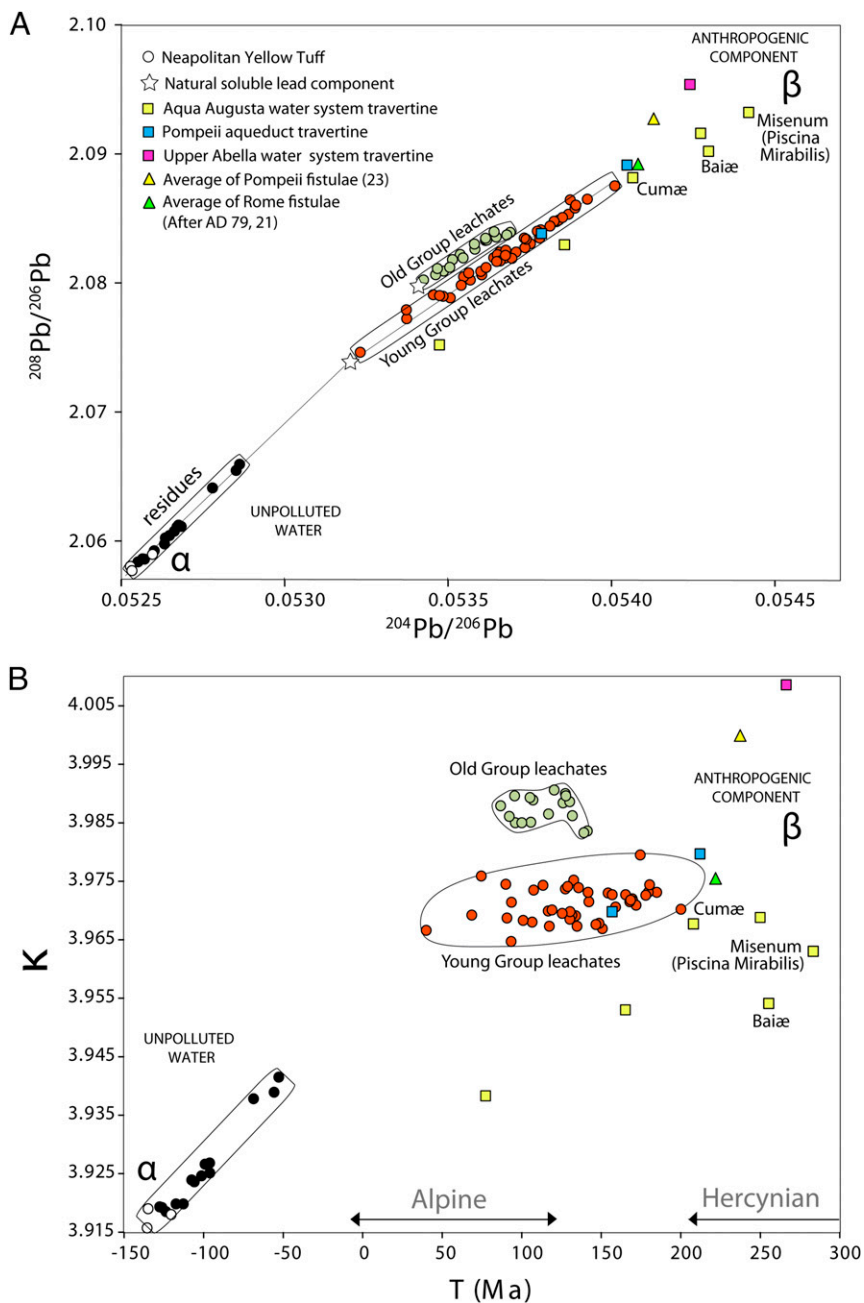


Fig. 3. (A) Plot of $^{208}\text{Pb}/^{206}\text{Pb}$ and $^{204}\text{Pb}/^{206}\text{Pb}$ for leachates and residues from the ancient Neapolis harbor deposits. Neapolitan Yellow Tuff (open circles), travertine (filled squares), and *fistulae* (filled triangles) (21, 23) are also shown. The residues define a mixing line between a volcanic component best represented by the Neapolitan Yellow Tuff and a natural fluvial (soluble) component represented by the star symbols. The leachates define two well-separated fields, which both can be accounted for by a mixture between a fluvial component and the imported (anthropogenic) component β . (B) Similar plot using the geochemically informed parameters κ ($^{232}\text{Th}/^{238}\text{U}$) and tectonic model age T_{mod} of the lead sources. This plot shows that the imported Pb component β is of Variscan (Hercynian, ~ 300 Ma) age: such values of T_{mod} are unknown in peninsular Italy, demonstrating that this component reflects massive contamination of the harbor by lead from the water distribution system. The two groups of κ values are distinct, which indicates that a new network of Pb *fistulae* was installed in the wake of the Somma-Vesuvius AD 79 eruption.

geological provinces where ore deposits are mined. In Europe, T_{mod} closely maps the distribution of its Alpine, Hercynian, and early Paleozoic provinces. All of the points falling on both leachate arrays in Fig. 3A have high $^{232}\text{Th}/^{238}\text{U}$ ($\kappa \sim 3.96\text{--}3.99$) (Fig. 3B). The Old Group mixing line includes deposits with T_{mod} values ranging from 90 to 130 Ma and high κ values (~ 3.99) (Fig. 3B), whereas the Young Group mixing line trends toward Hercynian Pb model ages (~ 250 Ma) and slightly lower κ values (Fig. 3B).

Comparison of Fig. 3A and B indicates that the radiogenic ends of the leachate arrays correspond to Variscan (Hercynian ~ 300 Ma) lead. Variscan tectonic units are unknown in central and southern Italy (with the exception of Calabria), which have been geologically shaped by the Miocene Apennine orogeny. The Pb component (β) (Fig. 3) is therefore necessarily exotic to the Neapolis area.

Impact of the AD 79 Eruption of Vesuvius as Revealed by $^{207}\text{Pb}/^{204}\text{Pb}$ and κ

The separation of the isotopic composition of the local vs. imported Pb components in the sediments is especially striking in Fig. 3B, which shows the κ parameter as a function of the apparent Pb model age. Factor analysis (Fig. S1) of bulk sediment element abundances identifies Pb as a loner with a large loading on the second factor and clearly separated from other elements indicative of human activity, notably Sn, Ag, and Cu. The particular status of lead is because of the fact that, like many Roman cities—and in particular nearby Herculaneum, Pompeii, Puteoli, Cumae, Baiae, and perhaps Misenum too (3, 17, 23, 24)—Neapolis received drinking water through a network of lead pipes. Because Variscan ages are essentially unknown in Peninsular Italy, the Variscan model ages of the anthropogenic component present in the sediment leachates document that contamination

originated primarily from the lead used for the *fulvae* of the local water distribution system (2, 3, 17), even if other lead artifacts may also have contributed to a lesser degree. Similar lead contamination of drinking (“tap”) water by the urban distribution system has been documented in ancient Rome (21) and Pompeii (17) as well.

With the possible exceptions of Pompeii and Herculaneum, all these networks were linked to the Aqua Augusta (2), but the distribution tank (*castellum divisorum*) diverting water to Naples has not been preserved. Masonry from the aqueducts themselves is unlikely to have contributed significant lead to the Neapolis harbor deposits. Considerable survey (reviewed in ref. 3) and geochemical analysis (17) have failed to find any remains of lead pipes or fittings within the main line channel of the Aqua Augusta or in the Bolla aqueduct, consistent with such fittings—known from other Roman aqueducts (25–27)—having been temporary (28) or removed later for recycling (29).

The Variscan T_{mod} and high κ values of component β at the radiogenic end of the leachate mixing lines (Fig. 3) clearly place the origin of the imported Pb in Western Europe (Spain, the Alps, France, Germany, and England) (Fig. S2). Whether β derives from a single provenance or a mix of different Pb ores is not clear, but the rather tight clustering around the mixing arrays argues for a stable source. The imported component β of the pre-AD 79 Neapolis harbor leachates is very similar to the average of the pre-AD 79 Pompeii *fulvae* analyzed by Boni et al. (23). The names of Campanian elite families dominate lead ingots from Cartagena from the second century BC to the first century AD (30). These and other ingots from shipwrecks map out a heavily trafficked route from the Cartagena/Mazarron and Rio Tinto mines to Puteoli—from where both Neapolis and Pompeii imported their lead—and Rome (31).

The Neapolis harbor deposits clearly show that the sharp change in $^{207}\text{Pb}/^{204}\text{Pb}$ and κ at -421 cm (Figs. 2 and 3) between the Old Group and the Young Group postdates the tephra unit of the AD 79 eruption of Vesuvius (between -485 and -436 cm) (Fig. 2), a defining event in the history of the Bay of Naples. Above two intermediate samples (at -421 and -429 cm) (Fig. 2), which simply reflect the effect of bioturbation, the transition is sharp (Fig. 2) and reveals a major shift in the source of the water flowing into the harbor.

The AD 79 Somma-Vesuvius eruption may have damaged the water supply system in different ways:

First, the ground uplift on the flanks of the volcano before the eruption may have deformed the slope of the channel of the section of the Aqua Augusta located on the Somma-Vesuvius, or broken the channel itself, necessitating its replacement, as was the case at Ponte Tirone (6).

Second, earthquakes may have caused damage. The Bay of Naples is not, in general, an area of strong seismicity. However, written testimony [Pliny the Younger, book 6, letter 20 (32, 33)] attests that earthquakes increased in frequency and intensity in the last days before the eruption. Statius [in book 4, chapter 8, verses 1–7 of the *Silvae* (34)] and Plutarch [in *Moralia* 398E (= *De Pythiae Oraculis* 9) (35)] describe damage to towns from the Neapolis area. Although damage to the aqueduct masonry of the Aqua Augusta channel is conceivable, lead is robust and malleable, enabling it to withstand earthquake damage. Hence, damage to the *fulvae* distribution system of Neapolis is unlikely.

Third, fine ashes emitted during the eruption must have entered the aqueduct through any open access shafts, clogging the *fulvae* system (3), which would have also received ash through any Pompeian-style water tower (6). Pipe systems as described by Frontinus (28) were clearly not designed for massive cleaning and therefore necessitated a full overhaul.

There is little doubt that after the disaster of the AD 79 volcanic eruption, the Aqua Augusta required major repairs and replacement of multiple lead pipe conduits. Assuming that sediment deposition rate did not change drastically over the period

of interest (~ 1 cm per year), the Pb isotope record shows that the old, likely damaged system kept bringing water to the harbor for about 15 y [the inferred time delay between the upper part of the AD 79 event layer (-436 cm) and the sharp shift in the Pb isotopic compositions of the leachates (-421 cm) (Fig. 2)] before a new *fulvae* network was completed and “switched on” and the old network decommissioned, which from a sedimentological perspective is essentially instantaneous. This record suggests that construction started on the Aqua Augusta immediately after its destruction to allow for such a rapid switch.

$^{206}\text{Pb}/^{204}\text{Pb}$: A Proxy for 500 y of Urban Change at Neapolis

A spectacular trend of decreasing $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ isotope ratios with time (Fig. 2) attests to a steady increase of the imported component, even through the AD 79 eruption, until -325 cm (the first half of the fifth century AD) (Fig. 2). The Vesuvius eruption nevertheless shows a shift of $^{208}\text{Pb}/^{204}\text{Pb}$ relative to $^{206}\text{Pb}/^{204}\text{Pb}$ (Fig. 2). This trend reflects the expansion of the *fulvae* system, either by expanding the network of pipes servicing existing areas or expanding the network to new areas (urban development). The end of this trend is contemporaneous with, and explained by, the final breakdown of the Aqua Augusta between AD 399 (the last mention of the aqueduct in a textual source, *Codex Theodosianus* 15.2.8) and AD 472 and the administrative and economic collapse in Campania accompanying the Visigothic (AD 410–412) and Vandal (around AD 455–463) invasions, plague (AD 467), and the next Plinian eruptions of Vesuvius (AD 472 and AD 512) (2, 3, 36–38). The resulting overall decline in imported lead shows a saw-tooth evolution, with two sharp reductions starting at -215 cm (the second half of the fifth century AD) (Fig. 2)—probably following the Plinian eruption—and -164 cm (the first half of the sixth century AD) (Fig. 2), coinciding with the sacking of Neapolis by Belisarius (AD 536) and then Totila (AD 542) during the Gothic Wars (39). It is quite remarkable that each sharp drop in the imported Pb component (*fulvae*) is followed by a slow relaxation marking the return of Pb-contaminated waters (Fig. 2), a clear sign that a reduced peri-urban water distribution system was brought back to use, perhaps consisting of lead pipes carrying rainwater or, in the low-lying areas of the town, the water of the Bolla aqueduct. The dramatic decreases show that these repairs were much slower and of more limited extent than those in the aftermath of the AD 79 eruption, reflecting the comparatively much weaker administration and resources of the fifth century Bay of Naples.

The last shift in Pb isotopic composition (Fig. 2) of the harbor deposits shows that an increase in Pb contamination occurred at the end of the sixth century AD. A stamped lead pipe dated to the seventh century, found in 2003 near the ancient harbor of Naples (40), records its donation by a member of the town’s elite (41), suggesting renewed attention to the water distribution system of Neapolis, occasioned by the expanding territory and power of the town and possibly an influx of inhabitants from neighboring declining towns (42).

Materials and Methods

We sampled the stratigraphic section of Neapolis’ ancient harbor at high resolution by taking a total of 61 samples (one sample every 9 cm). The samples were analyzed for Pb concentrations and isotopic compositions by, respectively, quadrupole inductively coupled plasma mass spectrometry (Q-ICP-MS) and multicollector inductively coupled plasma mass spectrometry (MC-ICP-MS) at the Ecole Normale Supérieure de Lyon (Table S1).

Pb Concentrations. Sample dissolution and other manipulations were carried out in a clean laboratory under laminar flow hoods. After sieving at $63\ \mu\text{m}$, aliquots of 100-mg sediment (fraction $< 63\ \mu\text{m}$) from the stratigraphic section were dissolved in a 3:1:0.5 mixture of concentrated double-distilled HF, HNO_3 , and HClO_4 in Savillex beakers and left on a hotplate at 120 – $130\ ^\circ\text{C}$ for 48 h, then evaporated to dryness. Perchlorates and any remaining fluorides were converted to chlorides by drying down with distilled 6 M HCl. The

samples in solution in 6 M HCl were all clear, attesting to complete breakdown of the sediments. The samples were redissolved in 2-mL concentrated double-distilled HNO₃, from which ~10% aliquots were further diluted to 0.5 M HNO₃ and to which internal standards were added (2 ppb In). Lead concentrations were analyzed by Q-ICP-MS (Agilent 7500 CX). The upper limit of the blank contribution was a factor of 100,000 smaller than the sample Pb contents.

Pb Isotope Compositions. Aliquots of 500-mg sediment (to ensure that the analyzed sample aliquots were representative of the actual samples) <63 μm from the stratigraphic section were weighed out into clean Savillex beakers. The labile or anthropogenic component of the Pb of the harbor sediments was extracted by leaching first with Suprapur chloroform, then with dilute double-distilled HBr (both leaching steps including ultrasonication, heating, and rinsing with distilled water). Lead was separated by anion-exchange chromatography using dilute double-distilled HBr to elute the sample matrix and distilled 6 M HCl to elute the Pb. In addition to separating Pb from the leachates of 61 samples, Pb was also separated from the residues of 14 of these samples selected such as to cover the entire span of the sediment

section. Before Pb separation for Pb isotopic analysis, the residues were attacked in the same manner as described above for elementary Pb concentration measurement. The amounts of Pb extracted from all samples were large (>1 mg) and orders of magnitude above the total procedural blank of ~20 pg Pb. Lead isotope compositions were measured by MC-ICP-MS (Nu Plasma 500 HR) with added Tl for instrumental mass bias correction and sample-standard bracketing using the values of Eisele et al. (43) for NIST 981.

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