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Title: A lead isotope perspective on urban development in ancient Naples

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Abstract

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 the ancient cities of the Bay of Naples (Neapolis). Written accounts on urbanization outside of Rome are scarce and the archeological record sketchy, especially during the tumultuous 5th and 6th centuries AD when Neapolis became the dominant city in the region. Here we show that concentrations and 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 5th c. AD, though vital repairs to this critical infrastructure were still carried out in the aftermath of invasions and volcanic eruptions.

Significance Statement

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 Neapolis' urbanization and later major conflicts at the end of the Roman period and in early Byzantine times.

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Neapolis: Water supply and volcanism

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 archeological 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 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, Baiae, and Misenum (2, 4). The total length of the aqueduct including its branches was approximately 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 5th 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 cm-sized. The availability of piped water at Pompeii, and more broadly at all 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 archeological evidence from Pompeii itself disagree as to whether the town was receiving any piped water shortly before the eruption, while 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 archeological excavation of the ancient harbor of Naples undertaken at Piazza Municipio by 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 archeological materials (9-13) with better precision than ^{14}C or OSL dating, and they record the history of the city during the first six centuries 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 pre-harbor layers have not been found. The lack of a pre-harbor unit has been attributed to the dredging of the bottom sediments during the late 4th c./middle 3rd c. 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 in the residues at -469, -461, and -453 cm (N49R, N50R, and N51R), notably as a dip in $^{206}Pb/^{204}Pb$ and a spike in $^{208}Pb/^{206}Pb$ (Fig. 2). In the very illustrative plot of $^{208}Pb/^{206}Pb$ vs $^{204}Pb/^{206}Pb$ (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 the lowermost layers up to sample N45 (-421cm), and the ‘Young Group’, which comprises all 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 $^{238}U/^{204}Pb$ (μ) and $^{232}Th/^{238}U$ (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 19-22). Lead model ages T_{mod} (in million years = Ma) are proxies for the tectonic age of the geological provinces where ore deposits are mined. In Europe, T_{mod} closely maps the distribution of its Alpine, Hercynian, and early Paleozoic provinces. All the points falling on both leachate arrays in Fig. 3A have high $^{232}Th/^{238}U$ (κ~3.96-3.99) (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), while 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 3B 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 $^{207}Pb/^{204}Pb$ 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 kappa parameter as a function of the apparent Pb model age. Factor Analysis (Fig. S1) of bulk sediment analyses 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 due to 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 *fistulae* of the local water distribution system (2, 3, 17), even if other lead artefacts 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 divisiorum*) 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 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/or 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 *fistulae* analyzed by Boni et al. (23). The names of Campanian elite families dominate lead ingots from Cartagena from the 2nd c. BC to the 1st c. 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 $^{207}Pb/^{204}Pb$ and κ at -421 cm (Fig. 2 and 3) between the Old Group and the Young Group post-dates 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.

- The ground uplift on the flanks of the volcano prior to 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).
- Earthquakes. The Bay of Naples is not, in general, an area of strong seismicity. However, written testimony ((32, 33), Pliny the Younger, book 6, letter 20) attests that earthquakes increased in frequency and intensity in the last days before the eruption. Statius ((34), in book 4, chapter 8, verses 1-7 of the *Silvae*) and Plutarch ((35), in *Moralia* 398E (= *De Pythiae Oraculis* 9)) 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. Damage to the *fistulae* distribution system of Neapolis hence is unlikely.
- Fine ashes emitted during the eruption must have entered the aqueduct through any open access shafts, clogging the *fistulae* 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 years (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 fistulae network was completed and 'switched on' and the old network decommissioned, which from a sedimentological perspective is essentially instantaneous. This suggests that construction started on the Aqua Augusta immediately after its destruction to allow for such a rapid switch.

^{206}Pb/^{204}Pb: A proxy for 500 years of urban change at Neapolis

A spectacular trend of decreasing ^{206}Pb/^{204}Pb and ^{208}Pb/^{204}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 (1st half of the 5th c. AD) (Fig. 2). The Vesuvius eruption nevertheless shows a shift of ^{208}Pb/^{204}Pb relative to ^{206}Pb/^{204}Pb (Fig. 2). This trend reflects the expansion of the *fistulae* 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 (c. 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 (2nd half of the 5th c. AD; Fig. 2) – probably following the Plinian eruption – and -164 cm (1st half of the 6th c. 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 (*fistulae*) 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 5th c. 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 6th c. 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, Q-ICP-MS and 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 μm, aliquots of 100 mg sediment (fraction < 63 μm) 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 °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 HNO_3, from which ~10 percent aliquots were further diluted to 2% HNO_3 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 < 2 percent of 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. Prior to 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. 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. (44) for NIST 981.

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Figure Legends.

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 panel **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 archeological excavation of the ancient harbor of Naples is located a few meters below current sea level in front of Piazza Municipio (**B**). On the left-hand side of the photo is seen 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 due to rise in sea level. Panel **C** shows an example of the harbor stratigraphic section investigated in this study and located in the eastern part of the excavation site.

Fig. 2. Downcore variations of ^{208}Pb/^{204}Pb, ^{206}Pb/^{204}Pb, and ^{207}Pb/^{204}Pb in leachates, ^{208}Pb/^{206}Pb in residues, and Pb concentrations. The Young Group consists of all the samples above layer N45, and the Old Group of all 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 archeological 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, Fig. 3), sedimentologically (light red shading), by specific sedimentological features (pumice stones), and archeologically, by consistent archeological dates (i.e. the 2nd and 3rd date from the bottom of the section). The parallel drift of ^{208}Pb/^{204}Pb and ^{206}Pb/^{204}Pb through time towards geologically old Pb reflects an increasing influence of pollution by the Pb pipe network (*fistulae*) and is a measure of urban development.

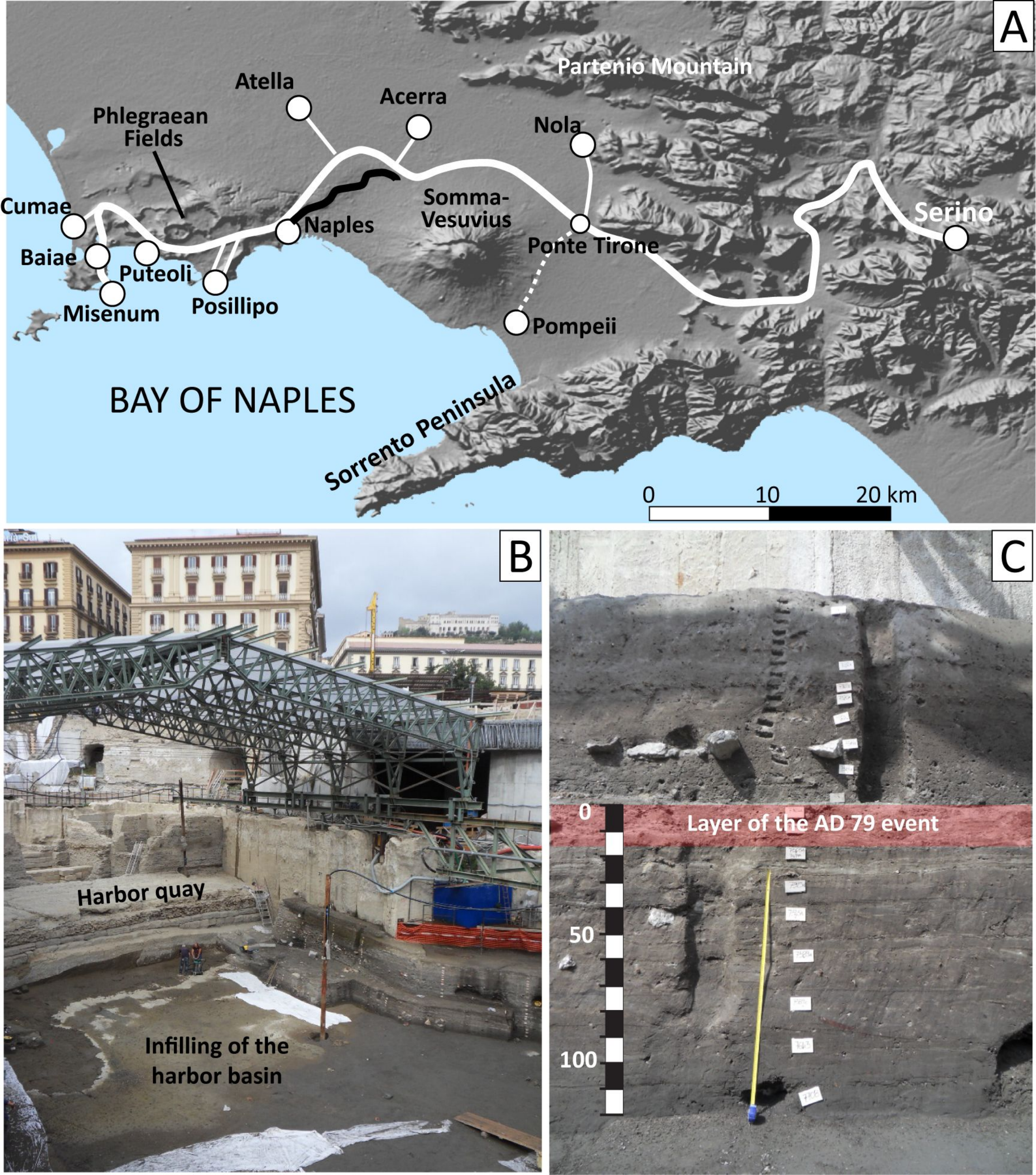
Fig. 3. (A) Plot of ^{208}Pb/^{206}Pb and ^{204}Pb/^{206}Pb for leachates and residues from the ancient Neapolis harbor deposits. Neapolitan Yellow Tuff (open circles), travertine (filled squares), and *fistulae* (filled triangles, 21, 23) are also shown. The residues define a mixing line between a volcanic 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}Th/^{238}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

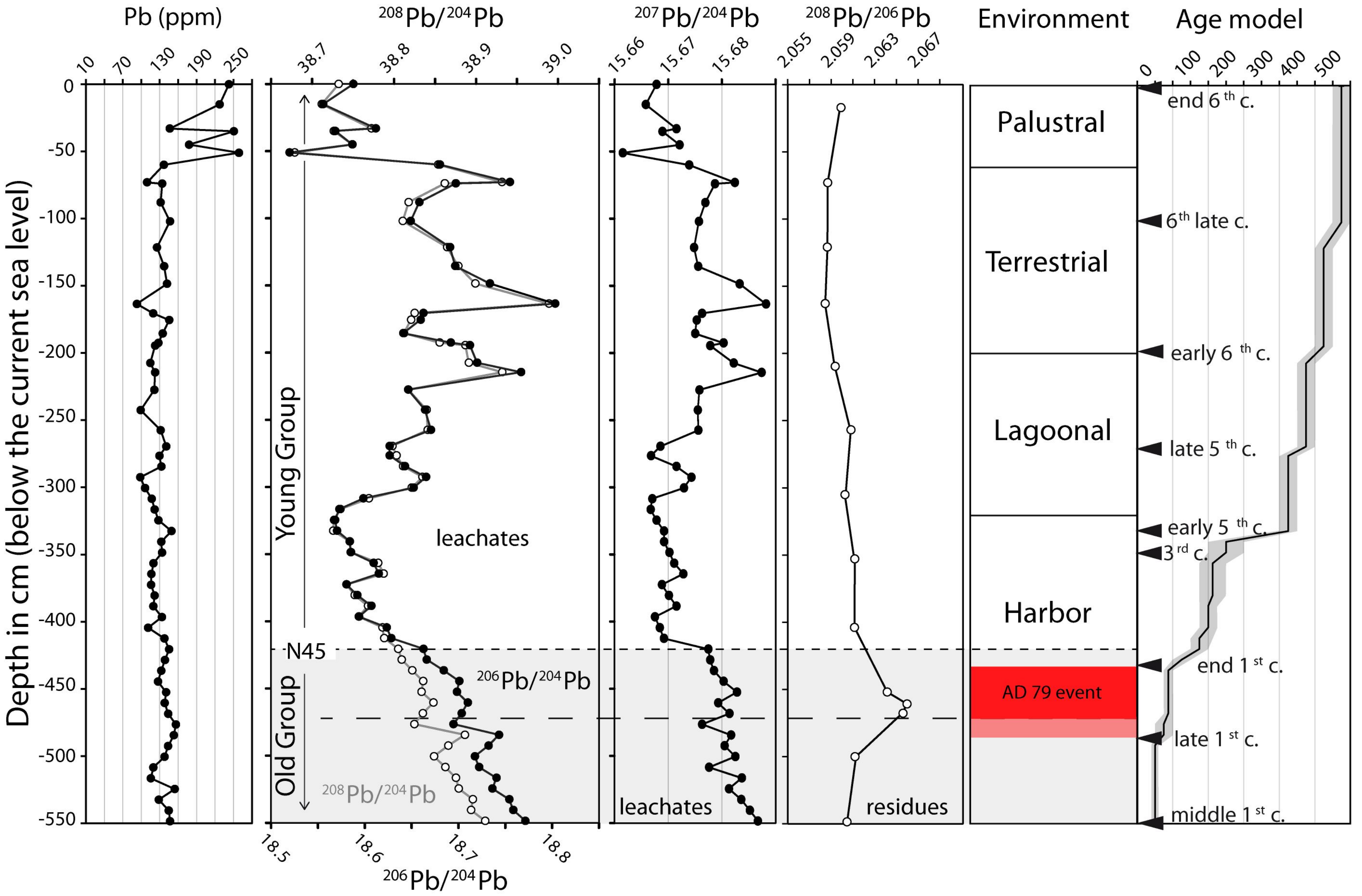
values of T_{mod} are unknown in peninsular Italy, demonstrating that this component reflects massive contamination of the harbor by lead from the water distribution system. The two groups of kappa values are distinct, which indicates that a new network of Pb fistulae was installed in the wake of the Somma-Vesuvius AD 79 eruption.

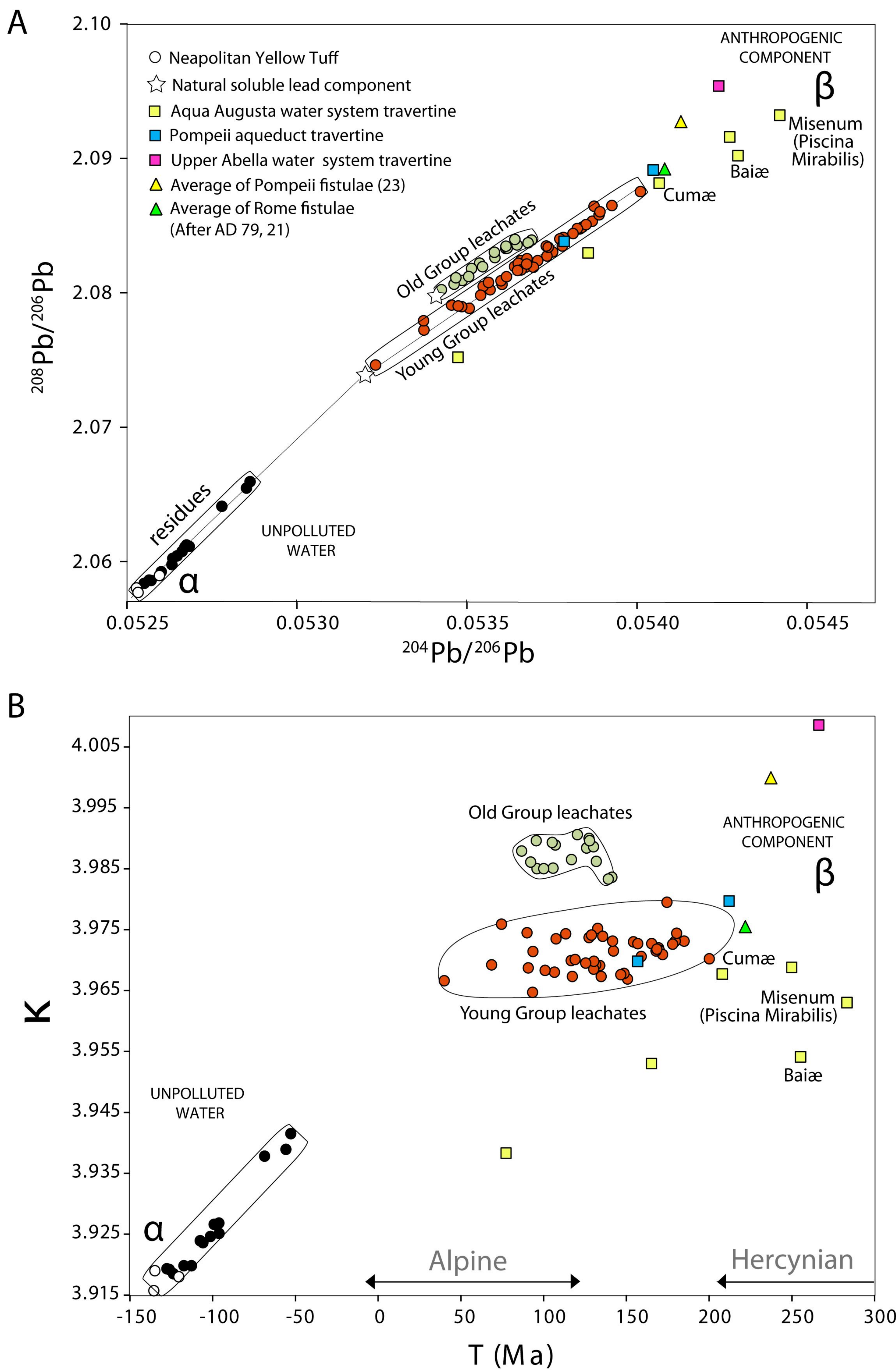
Any Additional Author notes.

Author contributions

H.D., J.B.-T., J.-P.G., and F.A. designed the research; H.D., P.R. and J.-P.G. did the field work and the sampling of the stratigraphic section; D.K.J sampled the water system travertines; H.D., F.A.G., and J.B.-T. produced the data; H.D., D.K.J., J.B.-T., and F.A. interpreted the data and wrote the paper


A
Partenio Mountain
Atella
Acerra
Phlegraean Fields
Nola
Cumae
Somma-Vesuvius
Naples
Serino
Ponte Tirone
Baiae
Puteoli
Misenum
Posillipo
Pompeii
BAY OF NAPLES
Sorrento Peninsula
0
10
20 km
B
Harbor quay
Infilling of the harbor basin
C
0
Layer of the AD 79 event
50
100


Pb (ppm)
10
70
130
190
250
^{208}Pb/^{204}Pb
38.7
38.8
38.9
39.0
^{207}Pb/^{204}Pb
15.66
15.67
15.68
^{208}Pb/^{206}Pb
2.055
2.059
2.063
2.067
Environment
Age model
0
100
200
300
400
500
Depth in cm (below the current sea level)
0
-50
-100
-150
-200
-250
-300
-350
-400
-450
-500
-550
Young Group
Old Group
N45
leachates
^{206}Pb/^{204}Pb
^{208}Pb/^{204}Pb
leachates
residues
Palustral
Terrestrial
Lagoonal
Harbor
AD 79 event
end 6 th c.
6th late c.
early 6 th c.
late 5 th c.
early 5 th c.
3 rd c.
end 1 st c.
late 1 st c.
middle 1 st c.
18.5
18.6
18.7
18.8
^{206}Pb/^{204}Pb


A
Neapolitan Yellow Tuff
Natural soluble lead component
Aqua Augusta water system travertine
Pompeii aqueduct travertine
Upper Abella water system travertine
Average of Pompeii fistulae (23)
Average of Rome fistulae
(After AD 79, 21)
ANTHROPOGENIC COMPONENT
β
Misenum (Piscina Mirabilis)
Baiæ
Cumæ
Old Group leachates
Young Group leachates
residues
UNPOLLUTED WATER
α
$^{208}Pb/^{206}Pb$
2.10
2.09
2.08
2.07
2.06
0.0525
0.0530
0.0535
0.0540
0.0545
$^{204}Pb/^{206}Pb$
B
K
4.005
3.995
3.985
3.975
3.965
3.955
3.945
3.935
3.925
3.915
Old Group leachates
ANTHROPOGENIC COMPONENT
β
Cumæ
Young Group leachates
Misenum (Piscina Mirabilis)
Baiæ
UNPOLLUTED WATER
α
Alpine
Hercynian
-150
-100
-50
0
50
100
150
200
250
300
T (Ma)