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# Production–consumption–release of trace metal elements, the urban proxies of the palaeo-anthropocene

Hugo Delile, Duncan Keenan-Jones

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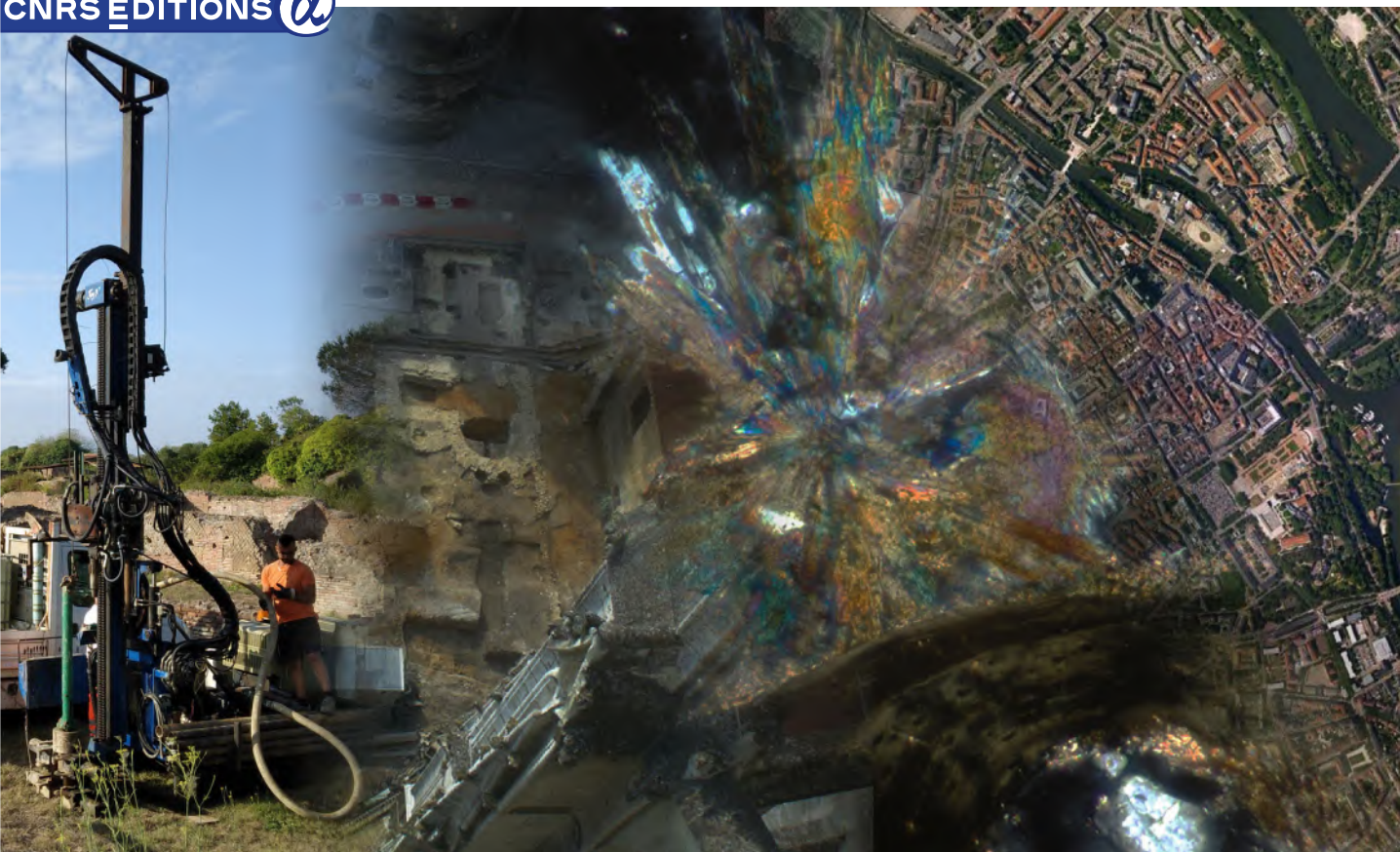
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Scientific editors:  
Quentin Borderie and Ferréol Salomon

# Urban Geoarchaeology

CNRS ÉDITIONS @



The global spread of urbanisation is having a profound impact on environments, climates and social lives. This gives rise to questions about the sustainability of urban areas, the management of soils and sediments and the socio-environmental construction of spaces. Archaeological investigations show that societies produced a wide variety of different urban forms throughout millennia and across continents. By focusing on human-environment interactions through the study of soils and sediments, geoarchaeological research provides new insights into the different ways of making and living in a city in the long term.

With twenty four contributions written by researchers from all over the world, *Urban Geoarchaeology* expands our knowledge of human-environment interactions and ancient urban lifestyles. This volume brings together substantial and comprehensive elements to understand past urban systems, using new interdisciplinary approaches, case studies of broad interest, and innovative techniques to answer historical questions. It shows how geoarchaeologists are producing and discussing new data on the transformation of landscapes through urbanisation. Processes of urban stratification over time are also better understood. The construction of urban spaces can be analysed from the large scale of the urban systems to the microscale of the soil formation, revealing ancient activities and lifestyles. Finally, *Urban Geoarchaeology* helps to connect the past, the present and the future of our cities.

Quentin Borderie is a field geoarchaeologist, based in Avignon (France), working on the urban environment for twenty years, giving courses at La Sorbonne University. He contributes to the research on urban Dark Earths, floor formation processes, and stratification dynamics in urban areas. He is attached to the CNRS laboratory UMR 7041-ArScAn, in the “Archéologies Environnementales” team.

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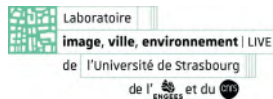
From left to right: Sedimentary drilling in Ostia (Italy) within the Roman city; archaeological excavation in Beauvais (France) from the top of the 16th century cathedral showing Dark Earths and an 11th century church; star shaped vivianite crystal and a bone through optic microscope cross-polarised light; aerial view of Metz (France) and the Mosel River.



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# Urban Geoarchaeology

Scientific editors: Quentin BORDERIE and Ferréol SALOMON



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# Production–consumption–release of trace metal elements, the urban proxies of the palaeo-anthropocene

Hugo Delile<sup>1</sup>, Duncan Keenan-Jones<sup>2,3</sup>

## Abstract

The image of the city in contemporary society has been viewed as the epitome of the Anthropocene epoch, during which damage to Earth system processes, the environment and human health have increased significantly since the mid-20th century (i.e., the 'Great Acceleration'). However, the dynamics driving the emergence, development and growth of cities have taken place over several millennia, and especially since the 1st millennium BCE when urbanization rates surged. Was environmental harm already perceptible in the earliest times of urban growth? And how can we detect it?

This chapter aims to address the hypothesis that the tripartite linkage between the production, consumption and release of trace metal elements (TME) could be used as a signature of the urban palaeo-anthropocene. Rather than a defined period of time, the palaeo-anthropocene could instead be seen as a conceptual framework in which the emergence and growth of urban societies are one of the key elements for understanding human exploration/expansion and landscape anthropization over a long time scale. Based on a non exhaustive overview of TME palaeo-pollution records from palaeo-environmental archives from Western Europe, South America and the circum-Mediterranean region, we find that the evidence converges on a relationship between the nature of the environmental archives investigated, the spatial extent of the palaeo-pollution signals, and different forms of anthropization. High-altitude and high latitude natural archives (i.e., glaciers, peatlands, and lakes) record the history of mining-driven atmospheric pollution from local to hemispheric scale, while contaminated coastal sedimentary infilling of ancient harbor basins documents long-term urban dynamics.

**Keywords:** Trace metal elements palaeo-pollution, palaeo-anthropocene, Pb isotopes, anthropization, urban water supply system, mining and metallurgy activity, South America, Western Europe, circum-Mediterranean region

## 1. Introduction

Since the pioneering work of Clair Patterson, the scientist who revealed lead (Pb) as a global problem in the environment and who spurred the movement against leaded gasoline, lead (Pb) releases, especially from leaded petrol and aviation fuel, acid batteries, electronic waste, paints, glazes and leaded glass, metal mining and smelting, and leaded water distribution systems, continue to be a major societal issue (WHO, 2021, 2022). Elevated blood lead exceeding the US Centers for Disease Control reference threshold (5 µg dL) appears to be widespread in low-income and middle-income countries (Ericson *et al.* 2021). Research on contamination by legacy micropollutants (trace metal elements [TME],

polychlorobiphenyls [PCB], polycyclic aromatic hydrocarbons [PAH]) and emerging micropollutants (pharmaceutical products) in the world's large river basins also converges on a relationship between chemical pollution-related public health risks and stages of economic development, with the highest anthropogenic pressures found in developing and industrializing low- to middle-income countries (Schwarzenbach *et al.* 2010, Shen *et al.* 2013, Delile *et al.* 2022, Wilkinson *et al.* 2022). Urbanization as a process of worldwide population concentration drives growing demand for energy and growing consumption of natural resources and is widely thought to be the primary source of environmental contamination and human health risks (Wilkinson *et al.* 2007, Schwarzenbach *et al.* 2010, Solecki *et al.* 2018, Delile *et al.* 2022). Cities have thus come to be seen as the environmental epitome of the Anthropocene epoch (i.e., the 'Great Acceleration'), as they play host to the agents of damage to Earth system processes (Steffen *et al.* 2015, Pincetl, 2017, Lade *et al.* 2020, Elmquist *et al.* 2021, Persson *et al.* 2022).

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Based on the geochemist Clair Patterson's pioneering work in the 1960s and 1980s, we now know that widespread atmospheric lead contamination of the northern hemisphere began as early as the first millennium BCE and peaked with the rise of the Roman Empire that brought large-scale lead silver mining activities (Patterson, 1965, 1971, 1972, Murozumi *et al.* 1969, Settle et Patterson, 1980). This mining output met the Roman Empire's needs for this metal, which found uses in urban, military, naval, domestic, funerary, medical, industrial, and even gastronomic applications, but it also exposed Roman period populations to lead poisoning by various channels (e.g., López-Costas *et al.* 2020, Burstein and Finch, 2022). Burstein and Finch (2022) performed a comprehensive inventory of literary texts referring to lead poisoning (citing a dozen authors: Nicander, Dioscorides, Scribonius Largus, Vitruvius, Pliny the Elder, Celsus, Galen, Aetius of Amida, Paul of Aegina, and Palladius) between the 2nd c. BCE and 7th c. AD that clearly shows how these ancient peoples had some knowledge of the human health effects of lead, but their understanding remained sketchy at best and far behind what we know today.

This statement of cities damaging environmental and human health for at least two millennia prompts the hypothesis that the tripartite linkage between production, consumption and release of TME, traces of which are recorded in palaeo-environmental archives, could be used as a tracer of the urban palaeo-anthropocene. Rather than a geological epoch, the palaeo-anthropocene could be seen as a conceptual framework capturing major contemporary issues raised by the 'Anthropocene epoch', such as global urban population growth, exploitation of natural resources, environmental and human contamination, and climate change. It represents an integrated holistic approach for understanding human exploration/expansion and anthropization processes over long timescales. The hypothesis supported here is that the increasing rate of urbanization from the 1st millennium BCE would be closely tied to the rising need for metals, and therefore, reflect the tripartite linkage between production-consumption-metal networks. This model implies that long-term trajectories for the emergence and development of cities increased the demand for metal ores, leading to an expansion of supply areas to new mining areas (local, regional or beyond). Exploitation of these ores and the resulting trade networks drove the emergence and development of new cities, thus further increasing the demand for metals. The tripartite linkage between production-consumption-metal networks suggests that the quest for and exploitation of metal ores have been key drivers for human exploration and anthropization of natural environments.

Here, to explore this assumption, we performed a nonexhaustive overview of the TME palaeo-pollution studies led in the three best investigated areas on this topic, i.e., Western Europe, South America, and the circum-Mediterranean region. For Western Europe, the studies selected focus on how the spatial extent of the TME palaeo-pollution signals depends on the nature of the environmental records investigated (high altitude glaciers, peatlands, and lake and fluvio-marine sediment deposits) and the tools and techniques used to identify mining sources of contamination. For South America, the selected studies highlight the diversity of palaeo-environmental archives used and their ability to track major technological changes in metallurgy. Finally, for the circum-Mediterranean region, the selected studies are based on fluvio-marine sediments and address emblematic issues such as the pre-anthropization of a settlement, long-term urbanization trajectories, and lead ore trade networks.

## 2. Recording TME palaeo-pollution: the issues of spatial scales, environmental archives, and contamination sources

### 2.1. Tracking long-term anthropization of natural environments and socioeconomic transformations in Western Europe

Clair Patterson's pioneering work in the 1960s and 1980s on the historical implications of metal production (Patterson, 1965, 1971, 1972, Murozumi *et al.* 1969, Settle et Patterson, 1980) spurred an entire field of research into past mining and metallurgical activity using environmental archives from Western Europe, mainly based on peatlands, lake deposits (i.e., Martínez Cortizas *et al.* 1997, Renberg *et al.* 1994, 2001, Shotyk, 2002, Le Roux *et al.* 2004, Kylander *et al.* 2005, De Vleeschouwer *et al.* 2007, García-Alix *et al.* 2013, Hansson *et al.* 2017, Allan *et al.* 2018, Elbaz-Poulichet *et al.* 2020, Corella *et al.* 2021), marine records (i.e., Leblanc *et al.* 2000, Martín-Puertas *et al.* 2010, Serrano *et al.* 2011, García-Alix *et al.* 2013) and Greenland ice (i.e., Hong *et al.* 1994, 1996, Rosman *et al.* 1997, More *et al.* 2017, 2018, Loveluck *et al.* 2018, 2020, McConnell *et al.* 2018, 2019, Preunkert *et al.* 2019). These studies have shown a continuous history of large-scale Pb atmospheric pollution in Europe over the past 3,000 years and earlier variations in natural background levels (Bindler, 2018). Note that much older phases of atmospheric TME pollution have been recorded in peatlands, especially in the earliest

and spatially narrower Chalcolithic/Bronze Age traces of mining and metallurgy in southeastern and southwestern Europe around 3600 BCE (Longman *et al.* 2018) and 3000 BCE (Martínez Cortizas *et al.* 2016), respectively.

The widespread Pb atmospheric pollution in Western Europe started in the early 1st millennium BCE, probably following the Phoenician western expansion motivated by lead-silver mining in the Iberian Peninsula (McConnell *et al.* 2018, Ben-Yosef, 2019, Eshel *et al.* 2019). The synchronicity around 1000 BCE between this initial phase of atmospheric metal pollution and significant global changes in land use (including the surge in rate of urbanization) evidenced by archaeological data (Stephens *et al.* 2019) shows how TME palaeo-pollution records provide a reliable indicator of landscape anthropization (Fig. 1). This relationship (illustrated in Figure 1) between urbanization and increasing atmospheric lead emissions from the beginning of the 1st millennium BCE could also intersect with the concomitant development of trade routes connecting mining districts to urban centers. F. Arnaud laid the groundwork for this relationship in the 2000s (Arnaud *et al.* 2006, 2010) advancing the hypothesis that lead-silver ores mined from the upper Arve valley in the Alps to supply the growth of the Roman city of Vienna was the source of the lead contamination recorded in the Lake Anterne during the Roman period.

The widespread and quasi-systematic traces of the major socioeconomic transformations recorded in peatlands, lakes and high altitude glaciers at high and middle latitudes is characterized by three phases of atmospheric deposition of anthropogenic Pb. Some scholars have called them the 'lead line', as they would represent a chronostratigraphic marker of past human activity (Fig. 1A) related to 1) the Roman lead-silver ore mining and smelting phase (~AD 0), 2) the Medieval lead-silver ore mining and smelting phase (~AD 1000–1200), and 3) the modern use of leaded gasoline (AD 1970) (Segard, 2009, Renberg *et al.* 2001, Alfonso *et al.* 2001, Wagreich and Draganits, 2018).

Recent advances in dating methods and Pb measurements in ice core archives at high resolution (cm resolution analyses) using inductively coupled plasma mass spectrometry (ICP-MS) and ultra high resolution (~120  $\mu\text{m}$  resolution analyses) laser ablation (LA)-ICP-MS provide annual and subannual Pb deposition time series, respectively (More *et al.* 2017, 2018, Loveluck *et al.* 2018, 2020, McConnell *et al.* 2018, 2019, Preunkert *et al.* 2019). Combining these geochronological models of continuous Pb measurements with geoarchaeological records and numismatic, archaeological (Loveluck *et al.* 2018, 2020),

historical and/or climate data (More *et al.* 2017, McConnell *et al.* 2018, 2019) has afforded reliable insights into the social, economic and environmental transformations of Central and Western Europe during the Late Holocene. For instance, the changes in the monetary and financial system (gold based to silver based coinage) of mid-7th c. AD northwestern Europe, probably driven by new, large-scale silver mining and bullion production areas, resulted in the increase in harbor settlements and trading in the Channel and North Sea during the second half of the 7th c. AD (Loveluck *et al.* 2018). Similarly, the signal of the mid-14th c. Black Death epidemic (bubonic plague) in Afro-Eurasia from AD 1346–1353 is well archived in ice layers from the alpine glacier Colle Gnifetti dated to AD 1349–1352 (Swiss-Italian Alps, More *et al.* 2017) and the Arctic AD 1349–1360 (McConnell *et al.* 2019) showing a significant drop in Pb concentrations following a sharp decrease in the number of European active mining areas in Europe at this time.

In Western Europe, there is ongoing debate over the long-distance transport of Pb from its sources (the lead-silver mining districts) into natural archives (polar ice caps, glaciers, peatlands and lakes), with the spatial extent of the recorded signals ranging from a macroregional or even hemispheric scale to a

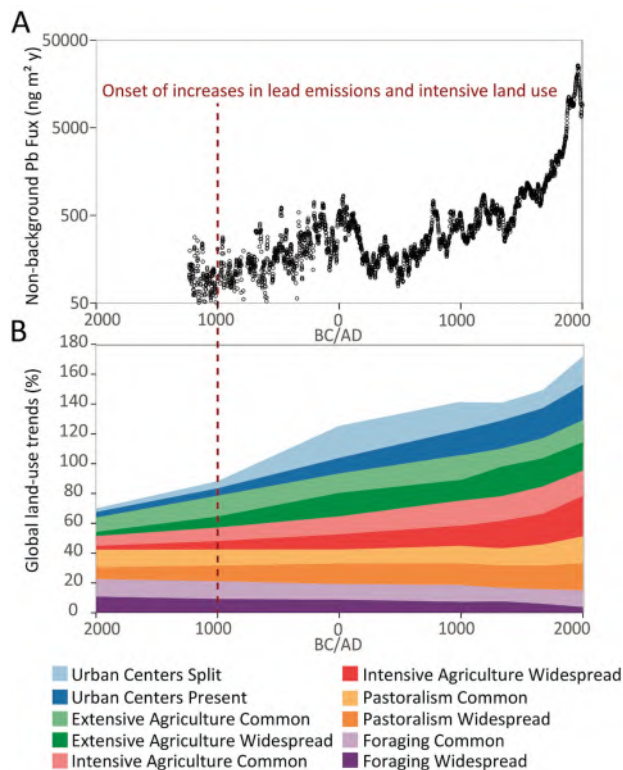


Figure 1: (A) Non-background lead flux recorded in a Greenland ice core (McConnell *et al.*, 2018, modified) and (B) global land-use trends (Stephens *et al.*, 2019, modified).

local and/or regional scale and showing strong spatial and temporal variability (More *et al.* 2017, 2018, Bindler, 2018, Silva-Sánchez and Armada, 2023). The advocates of an expanded scale make use of air mass movement research and modeling with *Climate Reanalyzer*<sup>™</sup> (More *et al.* 2017, Loveluck *et al.* 2018, 2020), *Flexpart* (McConnell *et al.* 2018, 2019) or *HYSPLIT* (Corella *et al.* 2021), while the advocates of a smaller scale refer to Pb isotope fingerprinting (e.g. Kylander *et al.* 2005, Mighall *et al.* 2014, Hinkley, 2018, Elbaz-Poulichet *et al.* 2020). According to Bindler (2018), the spatial spread of the Pb signal may range from local to regional scales depending on the nature of the environmental traps and their position in relation to the mining districts, but very rarely extends to a hemispheric scale.

## 2.2. Deciphering ancient metallurgical technology changes in South America

We have shown above that the terrestrial palaeo-environmental archives located at high altitude and high and middle latitudes (peatlands, lakes, and ice deposits) provide north hemispheric Pb pollution records spanning the last ~5,600 years, from which we can infer the history of European mining and metallurgy activity. Like for ancient Mediterranean civilizations, the economic and sociocultural development of successive Andean societies in South America was partly driven by the quest for rare metals (Delaere *et al.* 2019, Guédron *et al.* 2021). Ice, lake and peatland records have trapped atmospheric deposits of TME pollution released from mining and metallurgical activities by pre-Columbian civilizations within the Peruvian and Bolivian Andes that date back millennia (see references in Fig. 2 and Tab. 1). Starting in the 2000s, TME palaeo-pollution studies in the Andean region (Fig. 2 and Tab. 1) have focused on the onset and major stages (TME palaeo-pollution-related peaks) of the history of New World metallurgy according to pre-Columbian cultural periods (from Initial ~2000 BCE to Late Horizon, AD 1450–1532), Colonial times (AD 1532–1900), and the industrial era (20th c.).

The first archaeological evidence of copper (Cu) metalworking in South America dates back to the 2nd millennium BCE. The earliest Cu-Au artifacts were found in the central Andes at Mina Perdida (Peru) in excavations that dated hammered native Cu-based thin foils back to ~1400–1100 BCE (Burger and Gordon, 1998). South American Cu production by smelting began a few centuries later (Eichler *et al.* 2017), mainly in the Bolivian Altiplano, Northern Chile, and the Antofagasta and northeast Argentina regions (De Vleeschouwer *et al.* 2014). The first

atmospheric fallouts of TME released from copper metallurgy in the environmental archives are highly asynchronous (Fig. 2 and Tab. 1): the records span over nearly 3,000 years along a 4,700 km transect, with peaks of Cu and Sb trapped ~2000 BCE in the Karukinka peatland (Chile) (De Vleeschouwer *et al.* 2014) to the first Zn, Cu and Ag concentration peaks recorded in Laguna Pirhuacocha (Peru) at ~AD 1000 (Cook *et al.* 2007) (Fig. 2 and Tab. 1). The other lake records reviewed show an increase of Hg at ~1400 BCE in the Huancavelica region (Peru) (Cook *et al.* 2009a), Pb at ~AD 400 in Laguna Taypi Chaka (Bolivia) (Cook *et al.* 2008), and Cu, Ag, Pb and Hg at ~AD 900 in Lake Titicaca (Bolivia/Peru) (Guédron *et al.* 2021). South America's glaciers have also asynchronously recorded this initial atmospheric TME pollution phase, with an increase of Cu at ~700 BCE in the Illimani glacier (Bolivia) (Eichler *et al.* 2017) and Cu and Mo at ~AD 800 in the Quelccaya ice cap (Peru) (Uglietti *et al.* 2015). To compare with Western Europe, a recent study led by Elbaz-Poulichet *et al.* (2020) in sediment cores taken in Lake Robert (French Alps) shows that copper metallurgy began around ~1800 BCE, based on a first peak in Hg and Sb concentrations. Note that even allowing for the important margin for error in radiocarbon dating, the gap of nearly 1,000 years between the first local mining-related TME excesses recorded in Lake Robert and the earliest continental scale lead emissions trapped in Greenland ice could suggest that copper and silver mining outputs probably differed by several orders of magnitude.

Records marking the emergence of silver metallurgy in these different environmental records are also asynchronous to a degree, since evidence of silver metallurgy extends over 1,000 years along a 140 km transect with Pb excesses trapped around AD 500 in the Illimani glacier (Bolivia) (Eichler *et al.* 2015, 2017) to a Ag, Pb, Hg concentrations peak recorded in Titicaca lake (Bolivia/Peru) ~AD 1530 (Guédron *et al.* 2021) (Fig. 2 and Tab. 1). The other lake records reviewed show an increase of Pb, Sb, Bi, Ag, Sn ~AD 1000 in the Laguna Lobato (Bolivia) (Abbott and Wolfe, 2003, Cook *et al.* 2008), Pb ~AD 600 in the Llamacocha Lake (Peru) (Cook *et al.* 2009b), Pb, Sb, Bi in the Laguna Pirhuacocha (Peru) ~AD 1450 (Cook *et al.* 2007) and Pb, Sn in the Karukinka peatland (Chile) ~AD 1400 (De Vleeschouwer *et al.* 2014). Alpine lake records suggest that silver metallurgy began around 500 BCE, since the first peak of Pb concentrations occurred at this time (Tab. 1, Elbaz-Poulichet *et al.* 2020).

Note that according to these studies, the emergence of copper and silver metallurgy in South America ranges from 2000 BCE to AD 1000 and from AD 500

# TRACE METAL ELEMENTS, THE URBAN PROXIES OF THE PALAEO-ANTHROPOCENE

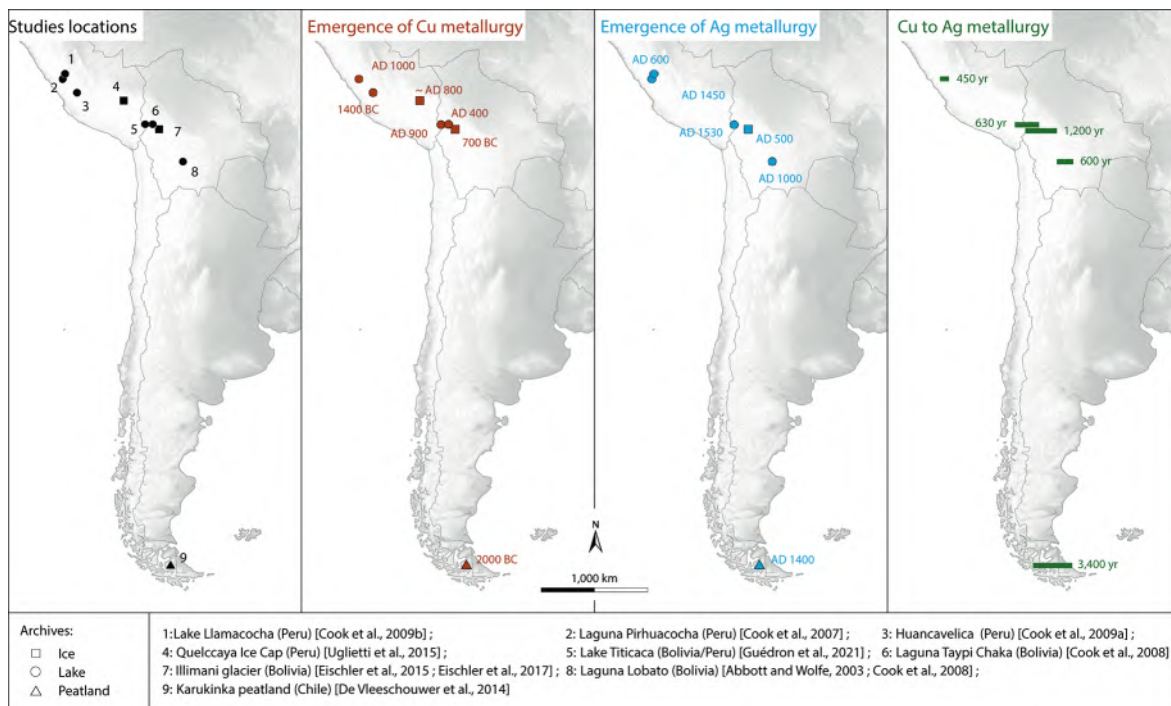


Figure 2: Emergence of Cu and Ag metallurgy in South America inferred from TME palaeo-pollution recordings into environmental archives.

Metallurgy	Element	Onset	Dominant cultures	Location	Geographic coordinates	Reference
Cu	Cu	700 BCE	Chiripa/Chapin	Illimani glacier (Bolivia)	-16.655767°; -67.783241°	Eischler <i>et al.</i> , 2017
Cu	Cu, Mo	~AD 800	Tiwanaku/Wari	Quelccaya Ice Cap (Peru)	-13.918797°; -70.814923°	Uglietti <i>et al.</i> , 2015
Cu	Cu, Ag, Pb, Hg	AD 900	Tiwanaku/Wari	Lake Titicaca (Bolivia/Peru)	-16.174407°; -69.024653°	Guédron <i>et al.</i> , 2021
Cu	Zn, Cu, Ag	AD 1000	Wari/Inca	Laguna Pirhuacocha (Peru)	-11.527027°; -76.082397°	Cook <i>et al.</i> , 2007
Cu	Pb	AD 400	Tiwanaku/Wari	Laguna Taypi Chaka (Bolivia)	-16.206623°; -68.351231°	Cook <i>et al.</i> , 2008
Cu	Hg	1400 BCE	Chiripa	Huancavelica (Peru)	-12.847222°; -74.952778°	Cook <i>et al.</i> , 2009a
Cu	Cu, Sb	2000 BCE	/	Karukinka peatland (Chile)	-53.888577°; -69.402361°	De Vleeschouwer <i>et al.</i> , 2014
Cu	Hg, Sb	1800 BCE	Bronze Age	Lake Robert (France)	45.131234°; 5.915171°	Elbaz-Poulichet <i>et al.</i> , 2020
Ag	Pb	AD 500	Tiwanaku/Wari	Illimani glacier (Bolivia)	-16.655767°; -67.783241°	Eischler <i>et al.</i> , 2015, 2017
Ag	Pb, Sb, Bi, Ag, Sn	AD 1000	Tiwanaku	Laguna Lobato (Bolivia)	-19.642249°; -65.696681°	Abbott and Wolfe, 2003
Ag	Ag, Pb, Hg	AD 1530	Colonial	Lake Titicaca (Bolivia/Peru)	-16.174407°; -69.024653°	Guédron <i>et al.</i> , 2021
Ag	Pb, Sb, Bi	AD 1450	Inca/Colonial	Laguna Pirhuacocha (Peru)	-11.527027°; -76.082397°	Cook <i>et al.</i> , 2007
Ag	Pb	AD 1000		Laguna Lobato (Bolivia)	-19.642249°; -65.696681°	Cook <i>et al.</i> , 2008
Ag	Pb	AD 600	Tiwanaku/Wari	Lake Llamacocha (Peru)	-11.082543°; -75.811234°	Cook <i>et al.</i> , 2009b
Ag	Pb, Sn	AD 1400	Inca/Colonial	Karukinka peatland (Chile)	-53.888577°; -69.402361°	De Vleeschouwer <i>et al.</i> , 2014
Ag	Pb	500 BCE	Iron Age	Lake Robert (France)	45.131234°; 5.915171°	Elbaz-Poulichet <i>et al.</i> , 2020

Table 1: Natural archives in South America used to trace the history of New World metallurgy

to 1530, respectively. Such time gaps also concern the transition from copper to silver metallurgy, which spanned about 1,300 years on a continental scale on average but with time gaps ranging from 400 to 3,400 years from one environmental archive to another (Fig. 2 and Tab. 1). These heterogeneous records of the main TME contamination phases could come from a number of factors, including the unreliable chronostratigraphic age models in South America (Giralt *et al.* 2008, Marsh *et al.* 2018, 2021). Indeed, the scarcity of terrestrial organic remains in lake sedimentary deposits forces researchers to resort to carbonate shells or aquatic plant macrorests for which the  $^{14}\text{C}$  reservoir effects are not firmly understood, which makes it hard to robustly calibrate of radiocarbon ages from this material (Marsh *et al.* 2018, 2021). For instance, lake water reservoir age estimates in Lake Chungará (Chilean Altiplano) fluctuate strongly, and Giralt *et al.* (2008) calculated a reservoir effect of 3,260 years, whereas Guédron *et al.* (2019) found a value 880 years older. For Lake Titicaca, Abbott *et al.* (1997) reported a reservoir effect of 250 years.

### 2.3. Lead palaeo-pollution trapped in the ancient harbor basins of the Mediterranean

One of the best studied environmental archives for TME palaeo-pollution purposes concerns fluvio-marine sediments deposited into ancient harbor basins around the Mediterranean, which were generally settled in deltaic environments (Tab. 2). This type of sedimentary deposit presents the specific feature of recording a much smaller spatial scale of contamination signal—generally associated with that of the city—than the other natural archives discussed above. Indeed, these TME pollution profiles are efficiently trapped in harbor sediments after being transported through the urban water distribution system, then sewers, and then the hydrographic network running through the city. Aerosol trace metals cannot be considered as a source of harbor sediment contamination as they carry very low concentrations of lead. For instance, in ancient times, especially the Roman period, the widespread atmospheric lead pollution caused an increase of about  $\sim 10 \text{ mg kg}^{-1}$  in Swedish lake sediments (Renberg *et al.* 2002) and a few  $\text{ng kg}^{-1}$  in Greenland ice (Fig. 1, McConnell *et al.* 2018, 2019). At the same time, records from harbor sediments captured several dozen to a thousand ppm above the geochemical background concentrations of Pb (Tab. 2). Such orders of magnitude of difference in Pb concentrations between environmental matrices point to a negligible contribution of aerosol trace metals to the Pb contamination of fluvio-marine sediments.

Nevertheless, lead contamination of harbor sediments during Antiquity remains significant (Tab. 2) since it is of the same order of magnitude as the in-sediment lead contamination measured at the river mouths of the current world's major rivers. Indeed, during the last 20 years, median values of particulate lead concentrations at large river outlets have ranged between 9 and  $81 \text{ mg kg}^{-1}$  (with highest values ranging from 37 to  $1885 \text{ mg kg}^{-1}$ ) (Delile *et al.* 2022).

Harbor geoarchaeologists have been studying anthropogenic lead records trapped in fluvio-marine sediments for the last twenty years, since they constitute a reliable and suitable proxy of site scale anthropization (Goiran and Morhange, 2003, Marriner and Morhange, 2006). In some cases, this geochemical tracer was able to identify phases of human occupation predating the foundation of the study sites. This was the case for Alexandria in Egypt where the first lead excesses measured in sedimentary deposits of Maryut lagoon and Alexandria Bay were dated to the first half of the 4th millennium BCE. This evidence of metal use on the Western margin of the Nile Delta during the Predynastic period at 3800–3500 BCE challenges the commonly accepted hypothesis of an urban creation *ex nihilo* of Alexandria in 334 BCE by Alexander the Great (Goiran, 2001, Véron *et al.* 2006, 2013). To date, this lead pollution is the oldest ever reported (Tab. 2).

Because of its low melting point and relative softness, lead was extensively used by all past human societies, and especially Roman populations who employed it in all areas of society. As a result, studies conducted on the TME palaeo-pollutions released by ancient cities have considered metallurgical activities as the main source of contamination of ancient harbor sediments (Tab. 2), and thus by extension consider this geochemical proxy as able to track past industrial activities (Le Roux *et al.* 2002, 2003, Véron *et al.* 2006, 2013).

## 3. Tracking long-term urbanization dynamics with harbor core Pb isotopic records

The recent association of the geochemical study of harbor sediments and archaeomaterials has recently made it possible to uncover a much more precise and reliable narrative of urban development in ancient cities, and even in some cases filling gaps in the knowledge when written accounts are scarce and the archaeological record is sketchy on past urbanization. Lead concentrations and isotopic compositions measured in sediments of ancient harbor basins, lead pipes (*fistulae*) and water system travertines show that

Region	Site	Pb concentration range (mg kg <sup>-1</sup> )	Onset of pollution	Source of pollution	Primary lead-ore supply source	Secondary lead-ore supply source	Reference
Eastern Mediterranean	Sidon (Lebanon)	5–70	1500 BCE	Metallurgical activity	Chyprus for the pre-Roman period	Rio-Tinto and Cartagena for the Hellenistic-Roman periods	Le Roux <i>et al.</i> , 2002, 2003
Eastern Mediterranean	Ephesus (Turkey)	2–241	Roman period	/	Cyprus and Dacia for the Roman period and the Early Middle Ages		Delile, 2014
Eastern Mediterranean	Ephesus (Turkey)	2–241	AD 1000	/	Thrace and/or western and northern Europe for the High and Late Middle Ages	Australian Paleozoic sulfides for the Modern period	Delile <i>et al.</i> , 2015
Southern Mediterranean	Alexandria (Egypt)	2–600	330 BCE	Metallurgical activity	Laurion and Siphnos (Greece) for the Greek period	Turkey for the Roman period	Véron <i>et al.</i> , 2006
Southern Mediterranean	Alexandria (Egypt)	1–286	1000 BCE	Building construction and manufacturing of ceramic vessels	/	/	Stanley <i>et al.</i> , 2007
Southern Mediterranean	Alexandria (Maryut Lagoon and Alexandria Bay, Egypt)	5–14	3800–2300 BCE	/	Cyprus, Turkey (Black Sea region and Taurus Mountains) around 2900–2300 BCE	Crete, Turkey (southern Black Sea region), and Southeastern Arabia in the Persian and Oman Gulfs around 3500–3800 BCE	Véron <i>et al.</i> , 2013
Southern Mediterranean	Utica (Tunisia)	7–210	350 BCE	Local mining activity	Tunisian metalliferous ores	/	Delile <i>et al.</i> , 2019
Western Mediterranean	Marseille (France)	2–350	AD 0–200	Metallurgical activity	Spain and France	/	Le Roux <i>et al.</i> , 2005
Western Mediterranean	Fréjus (France)	13–102	350 BCE	Use of metal ingots and forges	The Alps, the Languedoc, Cyprus and the Cyclades	/	Véron <i>et al.</i> , 2018
Western Mediterranean	Portus (Italy)	12–350	AD 100	Urban lead-pipe water supply network	Southwestern Spain, the Massif Central of France, the eastern Alps, Eifel in Germany, the Pennines in England	/	Delile <i>et al.</i> , 2014a
Western Mediterranean	Ostia (Italy)	22–2101	200 BCE	Urban lead-pipe water supply network	Southwestern Spain, the Massif Central of France, the eastern Alps, Eifel in Germany, the Pennines in England	/	Delile <i>et al.</i> , 2017
Western Mediterranean	Naples (Italy)	93–259	AD 50	Urban lead-pipe water supply network	Western Europe (Spain, the Alps, France, Germany, and England)	/	Delile <i>et al.</i> , 2016

Table 2: TME palaeo-pollution studies focused on harbor sedimentary archives in the Mediterranean (including isotopic lead data).

the lead piping used in the water supply networks of Rome, Ostia and Naples were the only source of radiogenic Pb, which is the hallmark of urban pollution (Delile *et al.* 2014a, 2015, 2016, 2017). This section presents the two main factors controlling the long-term urbanization dynamics, based on lead as a proxy of urban expansion and contraction, namely natural disasters and geopolitical drivers.

### 3.1. Urban development in ancient Rome affected by geopolitical hazards

The inferred history of the lead contaminated waters of Rome was based on cores taken from harbor sediments in the maritime ports of Rome located on the Tiber delta at Ostia (core PO2 in Figs. 3, 4), which was the first harbor to serve Rome, and at Portus using sediments from the Trajanic and Claudius basins (core TR14 and PTXI-3 in Fig. 3, 4), and a sediment core from the channel connecting Portus with the Tiber (Canale Romano) (core CN1 in Fig. 3, 4). The narrative of the urban development of ancient Rome based

on lead pollution is split into 6 main phases (Delile *et al.* 2014, 2017).

*Phase 1.* At both Ostia and Portus (Claudius' basin), anthropogenic Pb contamination in the harbor sediments began in around the mid-second century BCE (see Pb pollution index  $f_{\beta}$  values for PO-2 and PTXI-3 cores in Fig. 4). This dating means that the commissioning of *fistulae* in water systems is older than we thought, since the first dated stamps on lead pipes do not appear in Rome until 11 BCE (Fig. 4, Bruun, 1991), and it also means there was a delay of about a century and a half between the introduction of Rome's aqueduct system in the late fourth century BCE (Blackman and Hodge, 2001) and the installation of a piped grid. The start of anthropogenic lead emissions in urban freshwaters is followed by a sharp increase in the Pb pollution index  $f_{\beta}$ , which is read as a sign of urban expansion involving an increase in the spatial extent and/or density of the lead pipe system. This contamination phase is consistent with the repair and expansion of the water supply networks of Rome and the commissioning of the Aqua Marcia (late 140s BCE) and Aqua

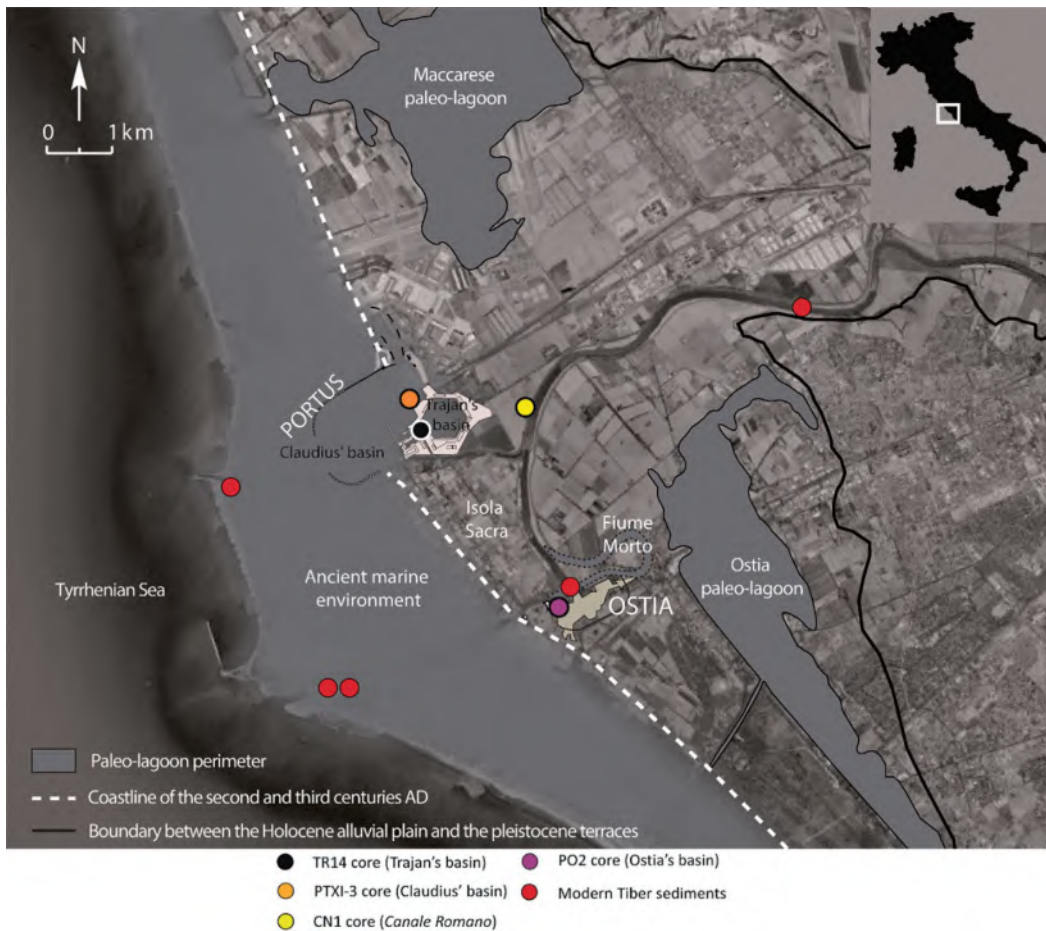


Figure 3: Location of the sedimentary cores and the modern Tiber sediments investigated in the Tiber delta (from Goiran *et al.*, 2014, modified).



Tepula (125 BCE) aqueducts (Blackman and Hodge, 2001) in order to meet the needs of Rome's growing population, which was likely tracking the growth in Roman citizens at this time (Fig. 4).

*Phase 2.* The PO2 core at Ostia showed a record of a sudden drop in freshwater Pb contamination during the first century BCE. This change occurred during a critical period for the Roman Republic, with civil wars that triggered a significant decline in the number of inhabitants and a sudden increase in the number of coin hoards (Fig. 4), and that slowed the construction of aqueducts and hampered their maintenance (Blackman and Hodge, 2001). This political instability thus translated into a halving of water flows in Rome's lead pipe distribution system, and on a larger scale into a sustained reduction of atmospheric lead emissions due to major interruptions of lead-silver mining in Western Europe (Fig. 4).

*Phase 3.* Once this period of unrest ended with Octavian and Agrippa's victory against their rivals in the early 30s BCE, the existing aqueduct and *fistulae* system were finally repaired, and three new aqueducts were built over the next 30 years (Bruun, 1991, Blackman and Hodge, 2001). This renewal of the urban water supply system during the Early Roman period is evidenced by a major spike in lead contaminated water in the Tiber River flowing into the harbor basin at Ostia (Fig. 4). This was followed by a shift in lead contamination from Ostia (decreasing Pb) to the Portus harbor sediments (increasing Pb) (Fig. 4). The most likely cause is a diversion of the contaminated Tiber water away from Ostia's harbor towards the new Portus harbor complex that was founded 3 km north of Ostia Antica in the middle of the 1st c. AD (Keay *et al.* 2005). The sustained anthropogenic Pb contamination of sediments during the Early Roman Empire recorded in multiple cores at Portus (cores PTXI-3 and TR14) and in the canal leading to them (core CN1) (Fig. 4) confirms the picture from textual accounts and archeological data showing that the aqueduct and lead pipe distribution system was generally maintained until at least the mid-3rd c. AD (Ashby, 1935, Bruun, 1991). At this time, the Roman Empire reached its apogee, especially in western European territories where extensive exploitation of ore resources (Domergue, 2008, Bode *et al.* 2009) resulted in the highest atmospheric Pb emissions (McConnell *et al.* 2018).

*Phase 4.* The next chapter in the history of Rome's lead contaminated water indicates a downturn in lead contamination during the Late Roman Empire (AD 250-500) that was recorded in the TR14 and CN1 cores at Portus (Fig. 4). This period of cleaner Tiber River water occurred at the same time as a new phase of geopolitical instability beginning in the 3rd c. AD (i.e., Gothic wars) until the 6th c. AD, and plunged Rome into the Imperial Crisis. This historical event

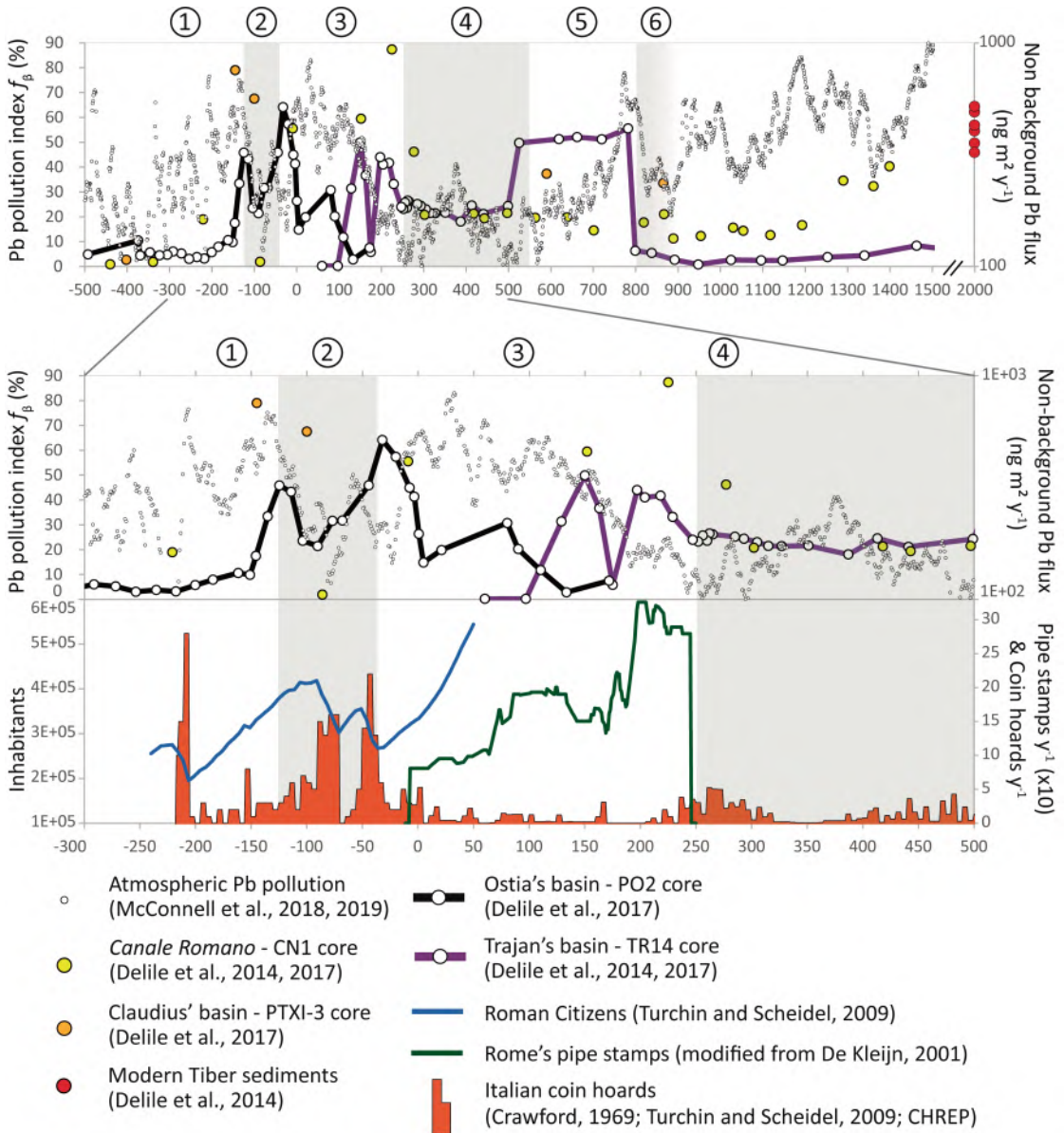
is well tracked on the Western European scale, since Greenland ice cores record an apparent decline of mining and smelting emissions (Fig. 4), thus confirming the picture of an overall economic downturn in the Later Roman Empire (Scheidel, 2009). In Rome, this troubled period is well tracked by a sudden increase in the number of coin hoards, the controversial decline in the population (Lo Cascio, 2001, Turchin and Scheidel, 2009), and an absence of pipe stamps (Fig. 4). Given that no more aqueducts were built and that maintenance was relatively minor at this time, the lower pollution of Tiber water could reflect a phase of contraction of the urban water supply system (Bruun, 1991).

*Phase 5.* During the Early Middle Ages (AD 500-800), there was a massive surge of lead in ancient Rome's city waters that was coeval with Byzantine repairs of the water distribution system at the end of the Gothic Wars (535-554 AD). At this time, massive amounts of corrosion products of decommissioned lead pipes could have been remobilized when Belisarius proceeded to recommission the aqueducts of Rome (Coates-Stephens, 1998). After a long period of reduced mining activity since the Late Roman Empire, aerosol TME emissions rose once again from Late Antiquity (Fig. 4).

*Phase 6.* There were no more anthropogenic lead excesses measured in sedimentary archives from Portus after AD 800. It is hard to identify an anthropogenic or natural cause for this disappearance of Pb contamination from the Tiber River, because at this period Portus was no longer functional after its basins and canals had silted up under floodplain deposits (Mazzini *et al.* 2011, Delile *et al.* 2014b). We can therefore speculate that a military event, such as the Arab sack of Rome in the mid-9th century destroyed the water distribution system (Duchesne, 1955) or that the increasing frequency of the Tiber's floods during this period (Aldrete, 2007) caused a dilution of lead content in the Tiber River.

### 3.2. Urban development in ancient Naples affected by natural hazards

Freshwater supply systems are key to urbanization in cities (Crouch, 1993, Kaniewski *et al.* 2017, Penny *et al.* 2018), and thus require detailed analysis of their resilience to geopolitical hazards, as seen above for Rome, and to natural hazards, as we will see here for Naples. One of the challenges faced by Naples (Neapolis) was to stabilize the Aqua Augusta aqueduct, which had been built between 27 BCE and AD 10 during the reign of Augustus (Keenan-Jones, 2010, 2013), and make it resilient to the hazards of volcanic eruptions from Vesuvius (Marturano, 2008,



- ① Commissioning of lead pipes and the Aqua Marcia and Aqua Tepula aqueducts in Rome's water distribution system
- ② Neglected urban water supply system during Civil Wars
- ③ Repairs, extending and maintenance of the existing urban water supply system during the Early Roman period
- ④ Contraction of the urban water supply system during the Late Roman period (Gothic Wars)
- ⑤ Byzantine repairs of the urban water supply system during the Early Middle Ages
- ⑥ Decommissioning of the urban water supply system during the Late Middle Ages (Arab sack and/or historical Tiber floods)

Figure 4: History of Rome's Pb-contaminated waters cross compared against historical and archaeological proxies, and atmospheric lead pollution of Europe.

Bruun, 2012, Filocamo *et al.* 2018). The Augusta was a regional network supplying eight or nine cities and numerous villas through multiple branches. To assess a little known side effect of these eruptions, and especially that of the eruption of Vesuvius in AD

79, lead concentrations and isotopic compositions were measured on a well dated sedimentary sequence from Neapolis harbor spanning the first six centuries AD (Delile *et al.* 2016a, b), and on carbonate sinters

sampled from the Aqua Augusta and Pompeii aqueducts and the upper Abella water system (Fig. 5).

The samples studied define three groups where the natural Pb component alpha ( $\alpha$ ), defined by residues and Neapolitan Yellow Tuff, is compared to the anthropogenic Pb component beta ( $\beta$ ) that is defined by the *fistulae* and travertine deposits (Fig. 5). This  $\beta$  component corresponds to the sources of lead contamination in harbor sediments that are at the interface of these two components on the plot. Like Rome, the anthropogenic Pb excesses trace back to the

urban freshwater supply networks. The harbor sediments show two distinct groups of samples based on whether or not they came before or after the eruption of Vesuvius in AD 79 (Fig. 6): the first group predates this natural disaster (green squares in Fig. 5), while the second postdates it (orange squares in Fig. 5). This change in Pb isotopic signature shows that the vast water supply system of the city and Bay of Naples was damaged by the eruption, and then had to undergo massive repairs to bring it back into operation. According to the sediment deposition rate of  $\sim 1$  cm

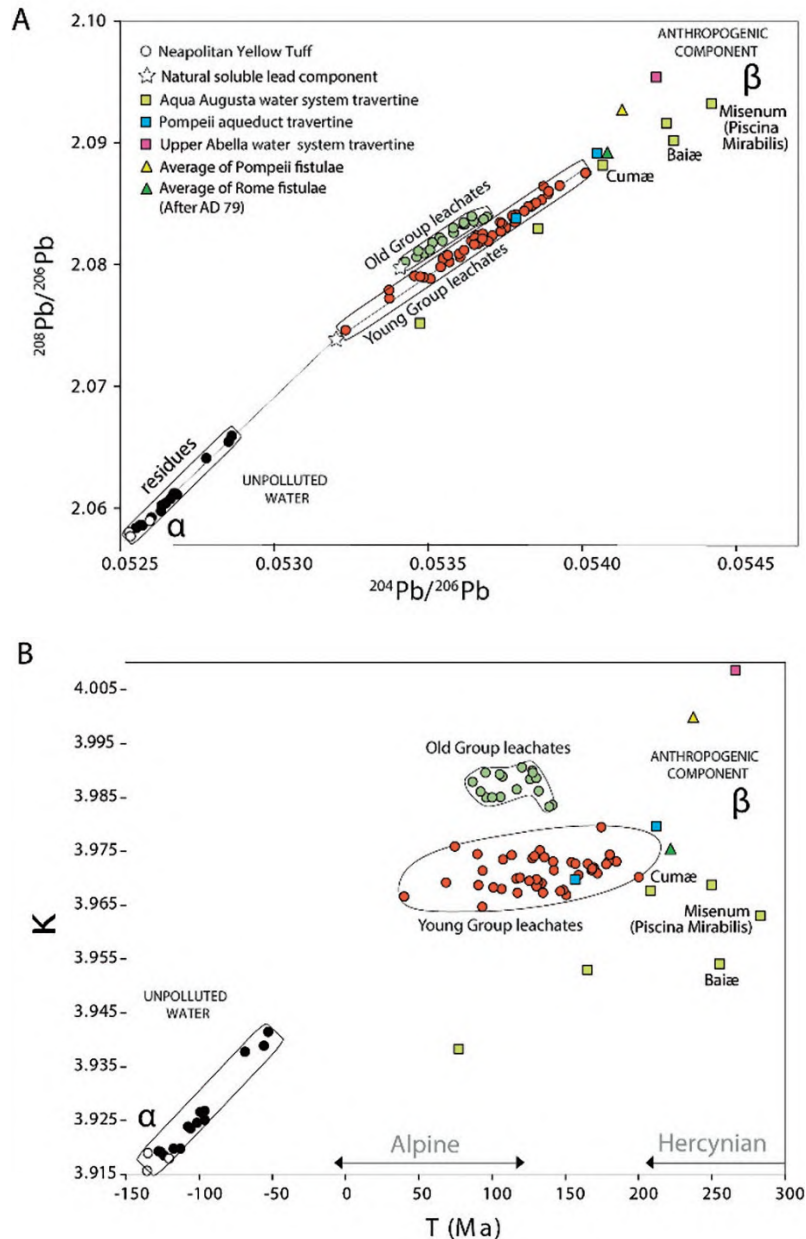


Figure 5: (A) Plot of  $^{208}\text{Pb}/^{206}\text{Pb}$  and  $^{204}\text{Pb}/^{206}\text{Pb}$  for leachates ('anthropogenic' Pb) and residues ('natural' Pb) from ancient Neapolis harbor deposits. Neapolitan Yellow Tuff (open circles), travertine (filled squares), and *fistulae* (filled triangles) are also shown. (B) Similar plot using the geochemically informed parameters  $\kappa$  ( $^{232}\text{Th}/^{238}\text{U}$ ) and tectonic model age  $T_{\text{mod}}$  of the Pb sources (from Delile *et al.*, 2016a).

per year, it appears that the old, eruption damaged system continued bringing water to the harbor for about 15 years before a new *fistulae* network was completed (Fig. 6).

Lead excesses trapped in the sedimentary archives of the ancient harbor basin of Naples also show isotopic discontinuities that translate different phases of the city's development.

**Phase 1.** From the 1st to the 5th century AD, the influence of the water supply system is increasingly manifest in the sediments through a pattern of sharply decreasing  $^{206}\text{Pb}/^{204}\text{Pb}$  and  $^{208}\text{Pb}/^{204}\text{Pb}$  isotope ratios with time (Fig. 6) that point to an expansion of the *fistulae* system, either by expanding the network of pipes servicing existing areas or by expanding the network to new areas (urban development).

**Phase 2.** From the second half of the 5th century, the period of urban development gave way to a period of urban contraction characterized by an overall decline in the anthropogenic Pb component  $\beta$  in sediments (Fig. 6). This phase is explained by the final decommissioning of the Aqua Augusta between AD 399 and AD 472 and the administrative and economic collapse in Campania driven by wars with the Visigoths (AD 410–412), Vandal invasions (around AD 455–463) and the capture of Neapolis by Belisarius (AD 536) and then Totila (AD 542), and by a disease epidemic (AD 467) and the next Plinian eruptions of Vesuvius (AD 472 and AD 512) (Soricelli, 2001, Savino, 2004, Cioni *et al.* 2008).

**Phase 3.** During the second half of the 6th century AD, the Pb isotopic composition of the harbor sediments show an increase in Pb contamination suggesting a fresh surge in urban development at Neapolis during the Early Middle Ages (Fig. 6). The fresh attention paid to the water supply network of Neapolis at this time is demonstrated by the discovery in 2003 of stamped lead pipe dated to the 7th c. AD near the ancient harbor of Naples (Carsana *et al.* 2005). This is probably a hallmark of the expanding power and territory of the city, and possibly an influx of inhabitants from neighboring towns in decline (Arthur, 2002).

## 4. Was the ancient urban growth embedded with large- or small-scale trade in lead–silver ores?

### 4.1. Tracing metal ore supply sources through lead isotope compositions

The method of tracing metal sources using Pb isotopes was introduced in the 1970s to determine the provenance of silver contained in Archaic and Classical Greek coins (Stos-Gale and Gale, 2009). It is based on the principle of traceability of geological sources in archaeomaterials, since each mineralization or metalliferous ore deposit has a specific Pb isotope composition that may be identifiable in metallic objects. This tool therefore requires an effort

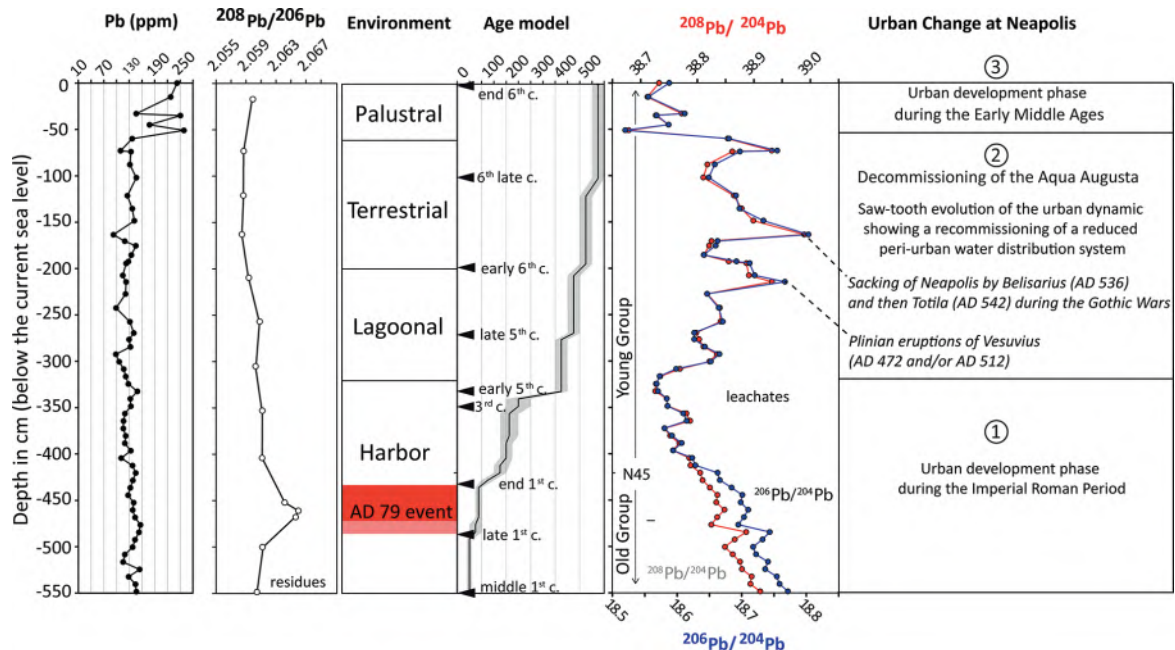


Figure 6: Downcore variations of  $^{208}\text{Pb}/^{204}\text{Pb}$  and  $^{206}\text{Pb}/^{204}\text{Pb}$  in leachates,  $^{208}\text{Pb}/^{206}\text{Pb}$  in residues, and Pb concentrations (Delile *et al.*, 2016a, modified).

to compare the Pb isotope compositions of metallic archaeomaterials with the Pb isotope compositions of potential geological sources of ores in order to establish a linkage. Performing this comparison thus requires merging individual Pb isotope datasets (the Oxford OXALID database, the national Swedish, Norwegian, and Finnish databases, and additional post 2001 literature based Pb isotope data) into one large geolocated database (Blichert-Toft *et al.* 2016, Madinabeitia *et al.* 2021, Klein *et al.* 2022, Tomczyk, 2022). Once this macro dataset has been built, it is possible to query the geolocated database to identify one or more correspondences to the Pb isotopic signal when studying objects and European and circum-Mediterranean ores (Delile, 2014, Delile *et al.* 2014a). The technique of using Pb isotope fingerprinting to identify lead ore supply sources is mainly limited by (i) overlapping of the isotope signatures of several ore deposit districts (e.g. Albarède *et al.* 2012, Artioli *et al.* 2020, Tomczyk, 2022), (ii) precision and accuracy of the reference data (e.g. Blichert-Toft *et al.* 2016, Klein *et al.* 2022, Tomczyk, 2022), and (iii) possible mixing of raw material (e.g. Eshel *et al.* 2021, Westner *et al.* 2020, Tomczyk, 2022).

#### 4.2. Lead ore supply sources for the Roman *fistulae*

Figure 7 reports results and findings from previous studies (Delile *et al.* 2014, 2016, 2017, Boni *et al.* 2000) on potential sources for the lead used in the *fistulae* of the water distribution systems in ancient Rome, Naples and Pompeii. Maps of Europe show pixels from the database that best agree with the raw data Pb isotope compositions of *fistulae* samples from these three cities. Squares correspond to potential sources for the lead used in the *fistulae*. The query on the database finds the pixels for which the error weighted distance to the mean *fistulae* composition in the Pb isotopes space is minimum. The color code corresponds to distances of 2 (red), 4 (yellow), 6 (green), 8 (cyan) and 10 (blue) sigmas, where one sigma equals 0.15 per mil of the isotopic ratio value.

Results show that the Pb *fistulae* of Rome were probably mined from ores in southwestern Spain, the Massif Central of France, the eastern Alps, Eifel in Germany, the Pennines in England, and Macedonia (Fig. 7). Factoring in the historical record of mining (Domergue, 2008, Bode *et al.* 2009) and the routes identified by shipwrecks loaded with lead ingots during the Late Republican Period and Early Roman Empire (Robinson and Wilson, 2011, Brown, 2011, Domergue *et al.* 2012), the most likely sources of lead are in the Spanish Sierra Morena, the English Pennines, the German Eifel, or the French

Massif Central. This kind of similar origin of lead ores contained in the *fistulae* of Rome, Naples and Pompeii is not surprising, as these three cities shared close cultural and commercial ties with each other and with the rest of Western Europe. Although the productive early mining areas of the Spanish Betic system were not identified as probable sources of the Pb in Rome's *fistulae*, most of the lead ingots from Cartagena from the second century BCE to the first century AD are stamped with names of elite Campanian families (Stefanile, 2015). Moreover, among the known maritime freight routes frequently marked by shipwrecks loaded with Pb ingots, the commercial harbor of Puteoli, one of the busiest centers of trade in the Roman Empire (Balland, 1965), connected the ore mining centers of Cartagena/Mazarron and Rio Tinto to the urban consumption centers of Rome, Neapolis and Pompeii (Domergue and Rico, 2014).

The fact that the sources for lead ore supply during this period were located in Western Europe suggests that the territorial expansion of the Roman Empire was driven partly by the need to secure new mineral resources (Wilson, 2007, Domergue, 2008), and thus that it was reaching its apogee. Given the relatively

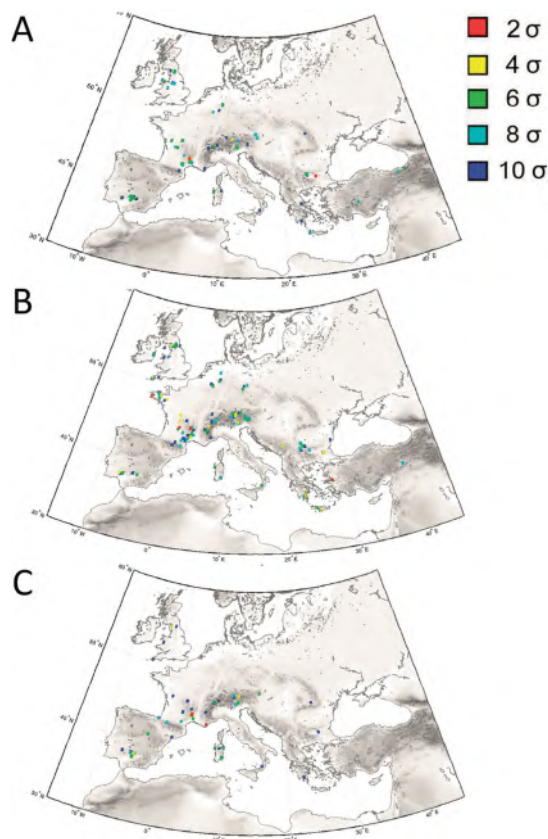


Figure 7: Potential sources for lead used in the *fistulae* of the water distribution systems in ancient (A) Rome, (B) Naples and (C) Pompeii.

short distances between the production and consumption centers, it is reasonable to assume that Rome, Neapolis and Pompeii dominated the economic spaces with which they were in direct contact. This regional pattern in the quest for mineral resources is also found in other Western Mediterranean port cities, such as Marseille, Fréjus, and Cala Francese where the lead ores used during antiquity were mined from the metalliferous Western European provinces of Spain (Cartagena and Mazaron, Rio Tinto, Arditurri) and France (Massif Central, Alps, Languedoc) (Tab. 2), as well as the major Eastern Mediterranean cities such as Ephesus, Sidon or Alexandria, where the lead ores identified in sedimentary archives were provenanced as imports from Greece (Laurion, Cyclades, Crete), Cyprus, Anatolia (Black Sea region and Taurus Mountains), the Middle East (Southeastern Arabia and the Oman Gulf) and Eastern Europe (Thrace, Dacia) (Tab. 2).

## 5. Conclusion

This non exhaustive overview of TME palaeo-pollution studies shows how TME patterns reported from different palaeo-environmental archives serve as a reliable proxy for tracing the geochemical impact of anthropization on air and water bodies at multiple scales of time and space. In both Western Europe and South America, high altitude and high latitude natural archives such as glaciers, peatlands and lakes record the history of atmospheric pollution from mining activities. In Europe, the signals trapped in these depositional environments show a striking chronological pattern of peaks that characterize three generalized phases of contamination called the 'lead line', i.e., a Roman phase (~0 AD), a Medieval phase (~1000–1200 AD), and a Modern phase (1970 AD), even though much older phases of atmospheric TME pollution induced by localized exploitation of mines have been recorded in European peatlands (i.e., 3600 BCE in Serbia and 3000 BCE in Spain). Conversely, in South America, records evidencing the emergence of copper and silver metallurgy through atmospheric fallouts of TME are highly asynchronous, as they span a period of nearly 3,000 years for copper and 1,000 years for silver. The spatial extent of the recorded signals related to the distance of TME transport from its mining sources is a topic of ongoing debate. In Western Europe, scholars defending a local and/or regional scale signal use Pb isotope fingerprinting to identify mining sources, whereas advocates of a much larger-scale signal (macroregional or even hemispheric) draw on air mass movement and modeling.

Efforts to date the emergence of copper and silver metallurgy in South America have so far found chronological gaps of a few millennia that span a 4,700 km transect for copper and a 140 km transect for silver, and also point to broad variability in the spatial extent of the palaeo-pollution signals recorded.

On the continental scale, recent ultra high resolution analyses of lead concentrations trapped in Greenland ice cores highlight an impressive synchronicity between the onset of increases in mining output from the beginning of the 1st millennium BCE and the concomitant global acceleration in the growth of influence wielded by urban centers. On a much smaller spatial scale, this relationship between TME palaeo-pollution and urban development is also evidenced in Mediterranean coastal sediments dating to antiquity. We showed that a more intensively targeted study of palaeo-pollution records taken in ancient harbor deposits, which are located at the outlet of urban hydrographic networks, can provide valuable insights into the dynamics and trajectories of long-term urbanization during the Greco-Roman period. Indeed, lead pollution resulting from the extensive use of lead pipe in water distribution systems, and trapped in harbor deposits, robustly traces the phases of urban expansion and contraction driven by natural and/or geopolitical factors. Based on the Pb isotope compositions of particle bound Pb excesses, the lead ore sources have been provenanced and shed new light on the metals trade networks and maritime routes used, through time and space, all around the Mediterranean basin. The large-scale trade in lead ores around the Roman Mediterranean, with sources from Western Europe, clearly shows how urban growth was deeply embedded with its trade networks.

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