

MOCIÓN QUE PRESENTA MARÍA DOLORES RUIZ ÁLVAREZ, CONCEJAL NO ADSCRITA DEL EXMO. AYUNTAMIENTO DE CARTAGENA, SOBRE “SUSPENSIÓN CAUTELAR OBRAS EN LA MORERÍA POR POSIBLE CONTAMINACIÓN DE SUELOS”

Esta concejal ha tenido conocimiento de que se ha presentado por registro municipal una denuncia en relación con las obras de adecuación del itinerario peatonal en Calle Morería Baja, en el entorno del Cerro del Molinete. El proyecto prevé movimientos de tierras, excavaciones y traslado de materiales, sin que conste una caracterización previa de los suelos afectados. Sin embargo, existen indicios científicos acreditados de contaminación histórica por plomo y otros metales pesados en el ámbito de actuación.

El proyecto estima que las tierras son no contaminadas, lo que podría dar lugar a su retirada y gestión como residuos inertes sin garantías sanitarias ni ambientales.

Esta situación supone un riesgo real e inmediato: Para la salud de trabajadores y población (exposición a polvo contaminado). Para el medio ambiente (dispersión de materiales contaminados). Para la legalidad en la gestión de residuos (pérdida de control y trazabilidad).

La normativa vigente obliga a identificar y analizar los residuos antes de su gestión, no después.

Por lo anteriormente expuesto la concejal que suscribe eleva al pleno la siguiente

MOCIÓN

Primero.- Instar la suspensión cautelar inmediata de cualquier movimiento de tierras en el ámbito del proyecto.

Segundo.- Exigir una caracterización ambiental previa de los suelos, que determine si contienen metales pesados y su correcta clasificación como residuo.

Tercero.- Prohibir la salida de materiales de la obra sin dicha caracterización y sin resolución administrativa expresa.

Cuarto.- Revisar el proyecto y sus estudios técnicos para adaptarlos a la normativa de residuos y prevención de riesgos

Cartagena, a 22 de abril de 2026

Firmado por MARIA DOLORES RUIZ ALVAREZ - DNI
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PROFESIONALES

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A LA ALCALDÍA – PRESIDENCIA DEL EXCMO. AYUNTAMIENTO DE CARTAGENA

Early metal pollution in southwestern Europe: the former littoral lagoon of El Almarjal (Cartagena mining district, S.E. Spain). A sedimentary archive more than 8000 years old

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Abstract A borehole drilling campaign has allowed the study of a former littoral lagoon located next to the harbour city of Cartagena in South-East Spain (close to the Sierra de Cartagena polymetallic ore deposits). This lagoon, which developed during the Holocene, was first a shallow sedimentary marine environment and later evolved into a swampy semi-endorheic basin named “Almarjal” (after the Arab term from the fourteenth century). The lagoon eventually dried out and at present forms part of the substratum of the modern sector of the city urban area. The basin representative sediments are sapropelic black silty facies forming a continuous sedimentary archive, accounting for more than 8000 years of depositional phenomena. The geochemical study of these sediments, together with their absolute calibrated dating by ¹⁴C, allows definition of successive stages of mining and metallurgical activities in the area. In turn, this information provides a more comprehensive perspective regarding metal pollution, particularly lead contamination during different periods of the Recent Prehistory and the Classical Age. The results indicate that the beginning of contamination by lead and other heavy metals occurred as early as 4500 years ago, when the Final

Chalcolithic period was taking place in the South-East of the Iberian Peninsula. This finding provides further insights regarding the debate on the origins of lead mining and metallurgy in SE Spain.

Keywords Heavy metal pollution · Holocene littoral basin · Lacustrine sediments · C-14 dating · Ancient mining and metallurgy · Cartagena (SE Spain)

Introduction

There are global evidences of atmospheric heavy metal contamination caused by the development of mining and metallurgical activities in early times, going back even to the Neolithic (Breitenlechner et al. 2010; Martínez-García et al. 2005; Mighall et al. 2002; Nieto et al. 2007; Nocete and coord. 2004; Nocete et al. 2005; Olías and Nieto 2015 and so on). This fact is especially important in the case of lead metallurgy, as it creates significant gas emissions transported through great distances due to atmospheric phenomena before their precipitation. The history of this contamination is especially well known in the case of Europe, thanks to the borehole study of the Greenland and the Arctic ice (Ferrari et al. 1999) together with the study of lacustrine and peatland sediments (Brännvall et al. 1997; Delgado et al. 2012; Olías et al. 2006; Pontevedra-Pombal et al. 2013; Shoty et al. 1998; Weiss et al. 1999; De Vleeschouwer et al. 2010, amongst other authors mentioned below).

In the case of those areas of study very far away of the early mining districts, the firstly detected significant increase of lead in the atmosphere belongs to the mining and metallurgical activities of the Roman period, followed by smaller rises during the Middle Ages and a significant increase starting at the Industrial Revolution (Weiss et al. 1999). In those areas

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closest to the early mining districts, such as the Mediterranean basins, we are finding out that the contamination by lead and other metals dates back to very earlier eras. We can find evidences in this regard for the French Mediterranean in the works of Le Roux et al. (2005), Miralles et al. (2006) and Elbaz-Poulichet et al. (2011).

Referring to Spain, the metal contamination has been studied in sedimentary records from different types of lacustrine basins and regions, on the Pyrenees (Camarero et al. 1998; Catalán et al. 2006; Oliva Urcia et al. 2013), on Sierra Nevada (García-Alix et al. 2013) and on the SW of the Iberian Peninsula (Borrego et al. 2002; Delgado et al. 2012; Olías and Nieto 2015). On the south of the Iberian Peninsula, the contamination has been studied as well with marine boreholes (Martín-Puertas et al. 2010; Mil-Homens et al. 2016; Rodríguez Bayona et al. 2012). More recently, metal pollution has been also studied at the La Molina peatland in Asturias (Martínez Cortizas et al. 2016).

In general, the basins more appropriate for the study of paleocontamination are those where the sedimentary series are preserved and without significant effects from post-depositional factors such as bioturbation, remobilisation, erosion and diagenesis, but this is not the kind of deposit that authors are studying, a transition coastal basin that evolved into continental with probable erosive or diagenetical processes. But in this case, the site selection has been based using other criteria; Carthago Nova, the most important Iberian-Punic city and port, was settled at this basin side. In the area, the largest economical activity was lead and silver metallurgy, and despite the sedimentary environment was not the best, the vicinity to the basin of the mineral-processing plants guaranteed the direct arrival of metal vapours/particles.

Anyhow, despite the differences amongst the different types of studied basins, a close correlation between the significance and increase of metal contamination and the mining and metallurgical development stages in successive cultural periods is always found (Martínez Cortizas et al. 1999; Nocete and coord. 2004; Nocete et al. 2005; Olías and Nieto 2015 and so on). In general, the geochemical record of these basins, including those very far away of mining districts, clearly shows the different episodes of metal contamination which took place in the Pre-Roman, Roman, Middle Ages and Modern eras. For example, the peak of Roman mining in South-Eastern Spain (Cartagena—the former Carthago Nova—and Mazarrón mining districts) clearly coincides with the lead relative maximum at 2200 BP standing out on the sedimentary record for the peatland of Penido Velloso in Lugo, very far away from South-Eastern Spain (Martínez Cortizas et al. 1999).

The comparison between the different studied basins that shows the importance of metal contamination varies especially depending on the type of basin (with or without contributions from drainage networks), the proximity to the emission sources and the importance of metallurgical activities (García-

Alix et al. 2013). Metal contamination has been especially important in the Western Mediterranean due to the proximity of significant metalliferous deposits at the Baetical area exploited from very early ages. In South-Eastern Spain, the origin of contamination has been dated at 3900 cal years BP (García-Alix et al. 2013); moreover, these authors detected that after 4500 cal years BP, there was an increase in the lead content, but they could not conclude whether it was human or natural induced. In South-Western Spain, the mining and metallurgical activities started in very early days at the Iberian Pyrite Belt deposits, whereas the beginning of metal contamination at the Guadiana river estuary dates back to 4500 years (Delgado et al. 2012). In North-Western Spain (La Molina, Asturias), the origin of the contamination has been recently dated back to an even earlier period, 4980 years ago (Martínez Cortizas et al. 2016).

The purpose of this study is to contribute to a better understanding of the origin and development of metal contamination in South-Eastern Spain through time, specifically in the area of the city of Cartagena. This was the former *chora* of the Punic *Qart-hadast* and the *territorium* of the Roman *urbs* of *Carthago Nova*, where the most important Pb-Zn deposits of the Western Mediterranean are located. Prior to this study, it was believed that these deposits were exploited from the Bronze Age and undoubtedly from the first millennium BC. For this purpose, we have used the data from the geochemical analysis and age dating by ^{14}C of the sedimentary record of eight boreholes by continuous coring. These boreholes were recently drilled in the former lacustrine environment of El Almarjal, adjacent to the city, as part of the multidisciplinary and comprehensive research “Carthago Nova: surveying and planning of a privileged Mediterranean City Arqueotopos I and II”, supported by the Ministry of Economy and Competitiveness of Spain and partially funded with FEDER grants (HAR2011–29330 and HAR2014–57672-P). Some of the goals of this research are the environmental and topographical retrospective of the city and its productive environment.

Regional setting

Geological context and mining history

The ancient city of Cartagena is located geologically on the southeastern tip of the Baetic Cordillera, in the so-called Internal Zone of the said chain. More precisely, it is located on the boundary between two major geological and geomorphological units, to the south the Sierra de Cartagena, a coastal mountain range, and to the north the Neogene-Quaternary plain of the Mar Menor or “Campo de Cartagena”, separated by a big fracture, the Cartagena-La Unión fault (Fig. 1) (Manteca Martínez y García García 2004). The Sierra de

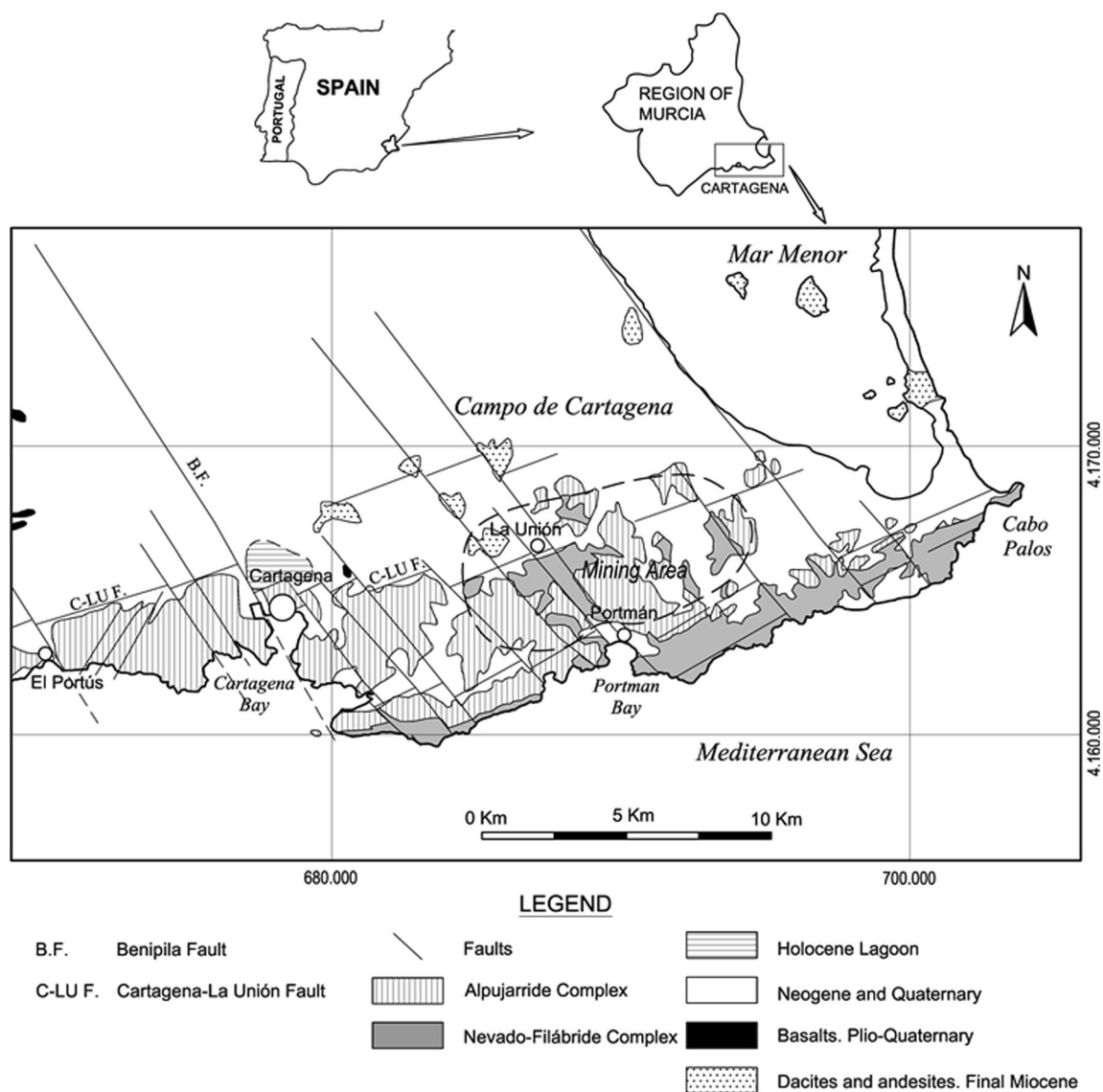


Fig. 1 Geological scheme of the Cartagena district (modified from Manteca and y Ovejero 1992)

Cartagena shows three mayor geological complexes superimposed on one another, from bottom to top, the Nevado-Filábride Complex (NFC) formed by metamorphic rocks, the Alpujarride Complex (AC) of slightly metamorphosed rocks and the Neogene (N) represented by a marine sedimentary formation of upper Miocene age. In turn, the Alpujarride Complex consists of two overthrust sheets, the lower one or San Ginés Unit and the upper one or San Julián Unit, both of Triassic age (Manteca y Ovejero 1992). To these sets, recent Quaternary sediments accumulated in depressions especially in the plain of the Mar Menor must be added.

These geological units are affected by volcanic calco-alkaline rocks (dacites and andesites) of Mio-Pliocene age (7–11 Myr) and alkaline basalts of Plio-Quaternary age (2.6 Myr) (Bellon et al. 1983). The morphology of the Cartagena mountain range and its littoral outline with its major

accidents, such as the Cartagena Bay and the Portman Bay (Fig. 1), are clearly influenced by the fault system, with its two main directions, N.70 and N.130. The intense hydrothermal activity linked to volcanic calco-alkaline stage allowed the formation of significant polymetallic deposits, mainly of Fe, Mn, Pb, Zn, Cu, Ag and Sn, that represents one of the largest Pb-Zn accumulations in the Iberian Peninsula (Oen et al. 1975; Manteca and y Ovejero 1992).

The main mineralisation are the “*mantos*” or strata-bound deposits, formed by replacement of Triassic limestones in the Alpujarride Complex (the “*primer manto*”) and of Triassic marbles in the Nevado-Filábride (the “*segundo manto*”) with a great lateral extension and thicknesses as much as 80 m in the case of the primer manto and 25 m in that of the segundo manto. There are also numerous veins with lengths between 500 and 1000 m and a metrical thickness. In addition, there are also deposit-type disseminations and stockworks; these latter

two types specifically affected Miocene sediments and volcanic rocks. There are also significant surface gossan-type deposits, formed by oxidation of sulphides. The original amount of all deposits of the Sierra de Cartagena has been estimated at 240 million tonnes (MT) of ore, containing as much as 64.5 MT of Fe, 3.2 MT of Pb and 3.8 MT of Zn and smaller amounts of other metals (Manteca and y Ovejero 1992).

This unusual concentration of minerals at Sierra de Cartagena has boosted an intense mining activity from the ancient days and during different historical periods. At the end of third and throughout the second millennium BC, consolidated copper-based mining and metallurgical activities were already taking place at this area of the Iberian Peninsula (Rovira et al. 1997; Montero-Ruiz y Murillo-Barroso 2010), although no archaeological evidences have been found. Even though during this period, the most wanted ores were those of copper, as they are closely linked to lead minerals on the deposits, copper metallurgy must have caused a significant lead contamination, which intensified somehow due to the use of lead as a replacement of tin for bronze metallurgy already at the end of the second millennium BC (García-Alix et al. 2013). All these facts may explain the presence of significant lead atmospheric contamination during these early days, as proven in Río Seco at Sierra Nevada and other locations (García-Alix et al. 2013).

There are documents proving the exploitation of complex copper ores containing silver and other metals such as grey coppers in Phoenician colonies on the Mediterranean and Atlantic regions of the Iberian Peninsula circa the eighth and sixth centuries BC. These grey coppers are linked to a copper-based metallurgy. Recent metallurgical findings in different locations of the Mediterranean region of the Iberian Peninsula, more specifically in the South-East, show us that the production of the Almería mining districts of Gádor and Sierra Almagrera and of the mining district of Sierra de Cartagena-Mazarrón were related to the said metallurgy (Renzi et al. 2009; Polzer 2014). Referring to this fact, we should highlight some studies of the origin of lead by isotopic characterisation that show the origin at the mining district of Sierra de Cartagena-Mazarrón for different metals and minerals used in other regions of the Iberian Peninsula, such as the South-East, the North-East and the Balearic Islands (Montero Ruiz et al. 2008). This fact is proven by the galena node and the cupel and litharge remains (group 3) found at the Phoenician colony of La Fonteta at the mouth of the Segura River (Renzi et al. 2009), as well as by the isotope matching the argentiferous mineral and cupellation remainders brought by the Mazarrón-2 vessel (idem 2009). Part of the load of this vessel came from the Phoenician metallurgical factory of Punta de los Gavilanes (Puerto de Mazarrón), which worked with argentiferous minerals of the mining district of Mazarrón during the seventh and sixth centuries BC (idem 2009; Ros Sala, e.p.). In this regard, recent data coming from a Phoenician vessel from the seventh to sixth

centuries BC sunk at Bajo de la Campana (close to Cabo de Palos, not far away from Sierra de Cartagena) show that already during the Phoenician era, there was a complex commerce of minerals of different origins and metallurgical treatments. The study of the mineral origins of the copper ingots, which were part of the load of this vessel, shows that they came from at least eight different mining areas of the Iberian Peninsula, including Sierra de Cartagena-Mazarrón (Polzer 2014).

However, it was clearly in Punic and Roman times when the mining activity in South-Eastern Spain and other areas of the Iberian Peninsula developed more intensely. The foundation of the *Qart-Hadash* city by Hasdrubal and its subsequent conquest by Scipio took place undoubtedly because of the benefits of exploiting the rich lead and silver deposits. Strabo (III, 2, 10), citing Polybius about the huge mining activity in Carthago Nova, explains that there were up to 40,000 men working in the area for this industry. Titus Livius (XXVI, 47, 6) described the Barcid city as “the most sumptuous in Hispania” because of its silver mines (Strabo, III, 4, 6). Several archaeological works confirm this huge exploitation and commerce of silver and lead minerals at the mining district of Sierra de Cartagena-Mazarrón (Domergue 1987; Domergue et al. 1990). To the many Roman lead ingots found at the harbour of Cartagena, we need to add those coming from other submarine findings at the nearby coast (Ramallo and Berrocal 1994; Ramallo 2011) as well as the Pre-Roman and Roman-Republican ingots found in different vessels and sites that were destinations of the Western Mediterranean commercial routes. The isotopic signature of these ingots together with the references of the vessel cartouches show that they come from the Cartagena-Mazarrón mining district (Trincherini et al. 2009; Bode et al. 2015). (Domergue, 1990 and Domergue, 2012)

After the Roman era, the mining and metallurgical activities on the area were occasional and not very relevant. It was during the middle-nineteenth century when the activity reached its peak again, thus marking the economic development of the region. Finally, mining on the area took place until the end of the twentieth century, with difficulties and after a long crisis following the two World Wars.

The core of all this huge industrial activity is in the central area of La Sierra, around 10 km to the east of the city of Cartagena, which is nowadays the town of La Unión (Fig. 1). However, a significant part of the metallurgic activity must have taken place very close to the city and its harbour. The mining activity causes a higher erosion rate and more metal contributions to the environment, especially to water via the associated drainage networks. In turn, the metallurgic activity brings metals to the atmosphere later added to the water and the ground. In this regard, the study of the sedimentary record of the El Almarjal basin gives us the chance for investigating the origin and evolution of these activities and their environmental consequences.

Location and description of the former lacustrine area of El Almarjal

This small littoral basin was first lacustrine and marine, and then, it evolved into a swamp that ended almost completely clogged between the sixteenth and seventeenth centuries (Conesa García y García García 2003). This basin was the northern border of the old city during the Pre-Roman and Roman periods.

This lacustrine area was the innermost section of an irregular cove conditioned by the fracture network (Fig. 2), with a continental narrowing in the centre connecting to the open sea to the south. As a result, there was a shallow lagoon landscape to the north and a double marine cove to the south, thus forming a peninsula on the area where the old city stood. In the Roman time, this inner part was nearly isolated from the rest of the cove due to the sedimentary contributions from the creeks (Polybius, X, 10, 12). Regarding this isolation in the second century BC, this author writes that “an artificial river-bed has been opened between the pond and the closest waters [...]”, which must have allowed the flow interchange between the lacustrine area and the bay. This fact proves that a great part of the perimeter of this sector was already a muddy environment. The historical cartography of this area from the Muslim era confirms the isolation of this inner section, which was a swamp or wetland.

Regarding the drainage, two main creeks flowed into the lagoon. The Hondón creek came from the east and drained a small sector of the Sierra de Cartagena mining area. The other was the Benipila creek, coming from the northwestern and much more important, which together with its tributary creeks of Los Dolores and Peñas Blancas drained a wide area but without ore deposits (Fig. 2). Consequently, the possible fluvial heavy metal contributions would reach the lagoon only through the Hondón creek.

Material and methods

Geological mapping of the Cartagena town area has been carried out to analyse the geological context of the El Almarjal basin. We have been able to define the basin morphology using the information obtained from a big number of boreholes, thanks to the collaboration of different companies specializing in geotechnical studies. Besides, now with the economic support of the General Department Technology of the Ministry of Economy and Competitiveness of Spain via the research project mentioned above, we have performed 19 new boreholes by rotation continuous coring, drilled specifically for the present study. Only 8 of those 19 boreholes are clearly located within the former lagoon. These eight boreholes, from 12 to 24.30-m deep, have been selected in order to obtain the

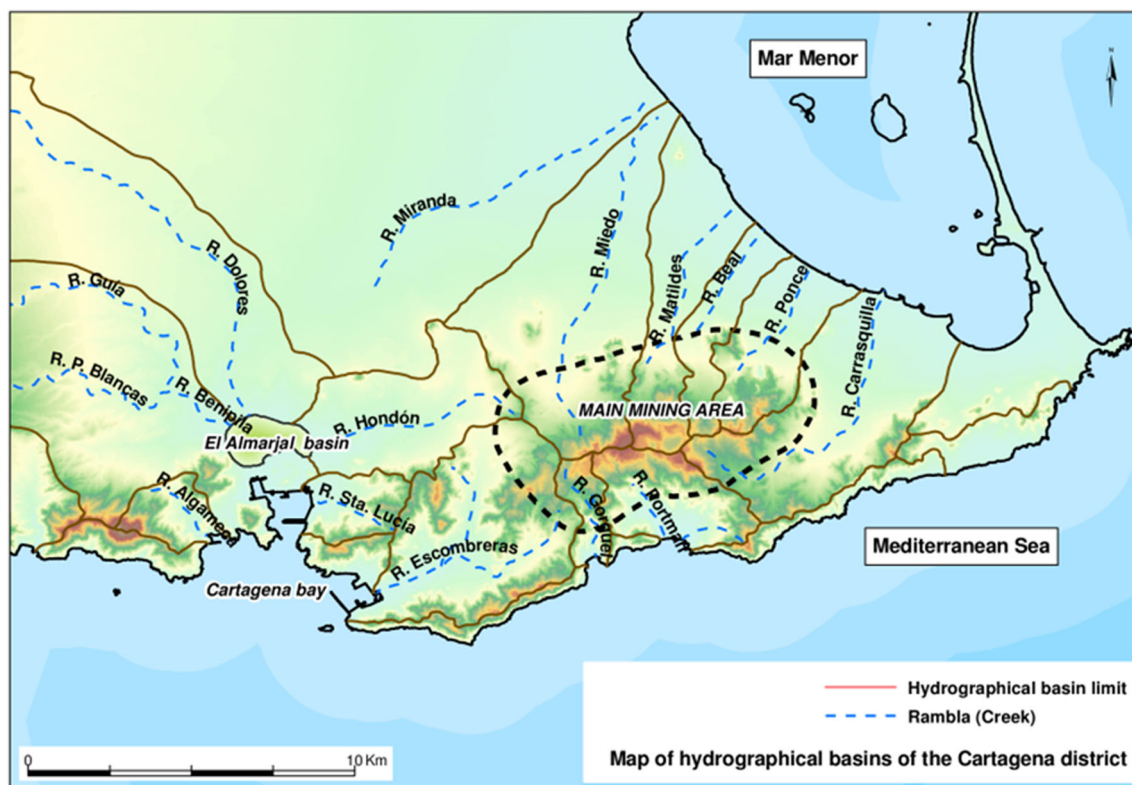


Fig. 2 Location of the El Almarjal basin in relation to the hydrographical basins of Sierra de Cartagena and its possible hydric connection with the mining area (current study) (topographic background DTM 1/25,000, Instituto Geográfico Nacional de España, 2015)

basin stratigraphy and its geochemical characterisation. Borehole sampling has been focussed on the swamp-lagoon facies sediments rich in organic matter (sapropelic silts). The samples correspond to 3-cm-thick core slices distributed at regular intervals along the boreholes (see sampling Table I in Annex). A total of 92 samples were taken to determine chemical composition of sediments. Moreover, we have analysed carbon isotopes from 32 of those samples in order to determine their radiometric age.

The chemical analyses were carried out at the Technological Instrument Service of Polytechnic University of Cartagena (SIT) using X-ray fluorescence spectrometry (XRF) with a Bruker S4 PIONEER machine. Before the analyses, the samples are prepared as appropriate by drying them out at 60° and grinding them using a mortar disc of size <100 µm. We then create a pill from each sample using 10 g of sample + 0.8 g of elvacite (measurement method Multares-Vac34; assessment method oxides-Vac).

A set of 32 radiocarbon datings by measuring the total organic carbon (TOC) in bulk sediment were carried out at the Poznan Radiocarbon Laboratory (Poland). All of them have been calibrated using version 7.0.1 of the Calib C14 programme (Stuiver, M. and Reimer, P.J. 1993; Reimer et al. 2013) (Table I in Annex). At first, in those samples with marine influence (406 and 407 cores), ages were corrected by using the “global” marine calibration curve Marine O4 (marine 13.14c) (Reimer et al. 2013) (in red in Table I in Annex). However, finally, we have adopted the ^{14}C calibrated values without marine correction, considering that the alkane analyses have determined that the vegetal origin of this organic matter is continental with marine influence, thus forming a brackish environment. The vegetal origin of the bulk organic matter can be feasibly determined through the alkane analysis. These organic compounds derived from plant waxes show long or short chain depending on the source plants, so in this case, the terrestrial environment has been clearly identified (Torres et al. in prep.) and no marine-alga contribution has been detected, thus making unable to apply marine correction. For a correct verification of the numeric datings obtained as per the International Union of Geological Sciences guidelines, we provide the lab and project sample ID and the sample depth above the sea level (m asl), together with the conventional ages (BP), the BP calibrated calendar medians (cal years BP), the calibration ranges to 1 and 2 σ and the different probability percentages for BC/AD dating estimation and the median (med. cal years BC/AD), which is shown with a minus symbol for the BC dating. We have systematically rounded up the different dating versions to the nearest ten from the value estimated by the Calib programme (Table II in Annex). Age-deep models were carried out using the software Clam.R. (v.2.2) (Blaauw 2010) on 403, 406, 407 and 417 cores. The interpolation system used was type 1 (linear interpolation neighbouring levels) within the linear function providing consistent results (Fig. 8).

To statistically correlate the chemical and chronological datasets and to compare the individual boreholes with the integrated model, an analysis of principal components (PCAs) has been performed with the statistical support of Research Support Services of the University of Murcia. The analysis has been carried out on the Pb, Cu, Zn and Al contents and depth sample variables; the age of samples has been taken into account as supplementary variable. The data have previously been standardised so that the coefficients calculated are correlations of each variable with the principal components. All data were processed using the R statistical software FactoMineR (Husson et al. 2015), version 3.2.2 (R Core Team 2015).

Results and discussion

Relative to the basin stratigraphy and morphology

The Almarjal lagoon deposits normally lie over Plio-Quaternary materials and sometimes over the Miocene or Baetic substratum (Figs. 3 and 4). When the basin peak, it was 1700-m long from east to west and around 700-m wide from north to south. The bathymetric profile of the sediment filling is asymmetrical, being smooth and wide on the north side and narrower and steeper on the south, conditioned by a N.70° fault (Cartagena-La Unión fault) delimiting the peninsula to the north. The south side of the basin is where the lagoon sediments are thickest (around 20 m), probably because of the subsidence of the sector in relation to the fault mentioned above. The coast limit of the west side of the peninsula formed a small cove between the actual hills of Concepción and Molinete (Fig. 4).

Its stratigraphic series, from the wall to the top, are as follows (Fig. 5):

- Basal section of gravels and brown sands around 10-m thick.
- Sapropelic sandy silts of dark grey colour and with abundant plant and shell remains, up to 12.6-m thick. The predominant shells are *Cerastoderma glaucum* and *Cerithium vulgatum*, together with other species typical from brackish transition environments. The sapropelic sediments indicates a very low dynamics and a reducing environment.
- On top of the sapropelic silts, we can find not very thick brown silty sands, whereas in other areas, we have anthropogenic infillings of different ages, dating back at least as early as the Roman era in some sectors.

The isobaths map created on the borehole data shows the existence of a strait around 300-m wide, which restricted the

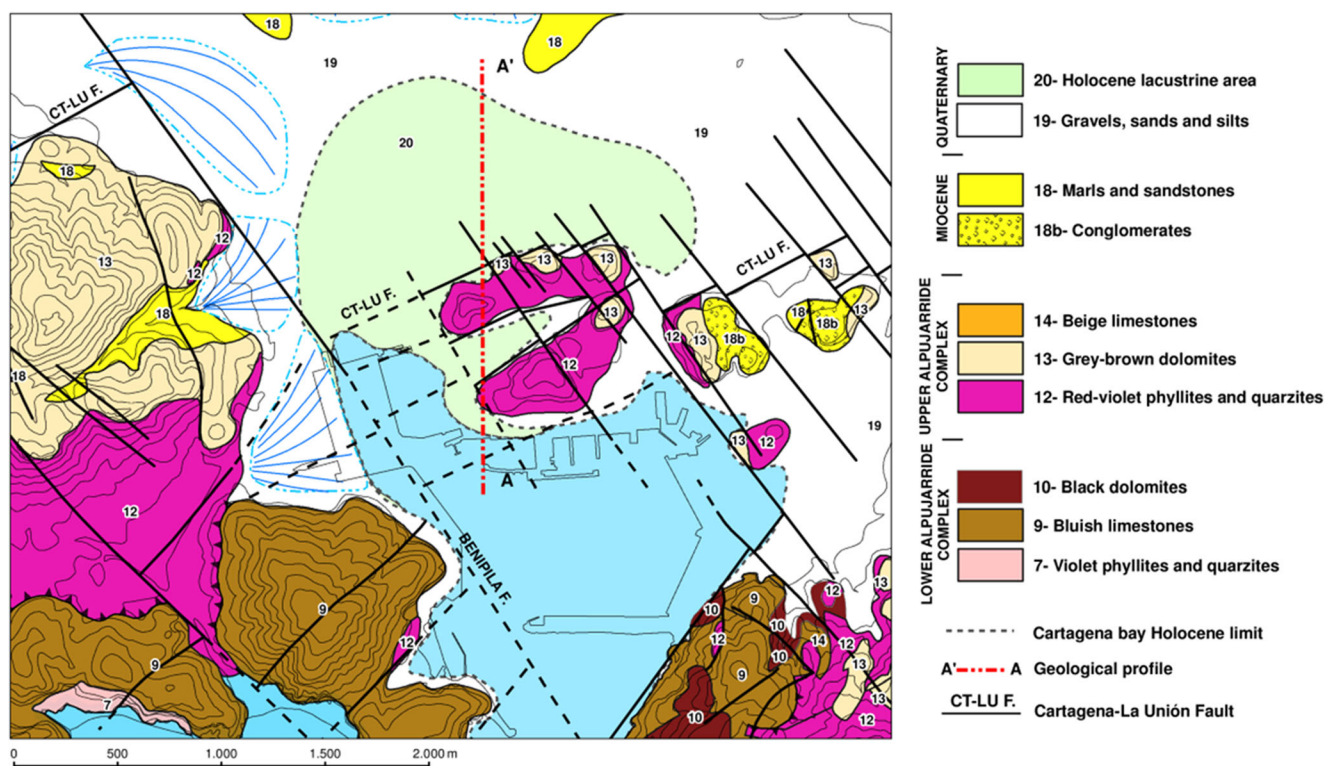


Fig. 3 Geological map of the Cartagena cove including the fault network conditioning its morphology and the representation of the lacustrine area of El Almarjal (current study, Arqueotopos project)

hydraulic flow with the bay and contributed to that reducing atmosphere (Fig. 6).

Relative to the preservation of the sedimentary records and their chronology

Regarding the preservation of the metal records, it is clear that the high organic matter content of the host sediments favoured the fast metal fixation, probably as organo-metallic molecules

that diffused very slowly. As well as other heavy metals, they can accumulate associated to organic particles forming films in a ratio of 10^4 when compared to inorganic ones (Duce et al. 1972). Besides, lead as vapour or particle easily reacts with carbonates forming cerussite (PbCO_3). In the Cartagena sedimentary record, the carbonate (calcite) is always present, thus ensuring that lead inerting as lead carbonate can be considered as total insoluble. In this way, the lead input results fixed following different ways and the detected ^{14}C heterochrony

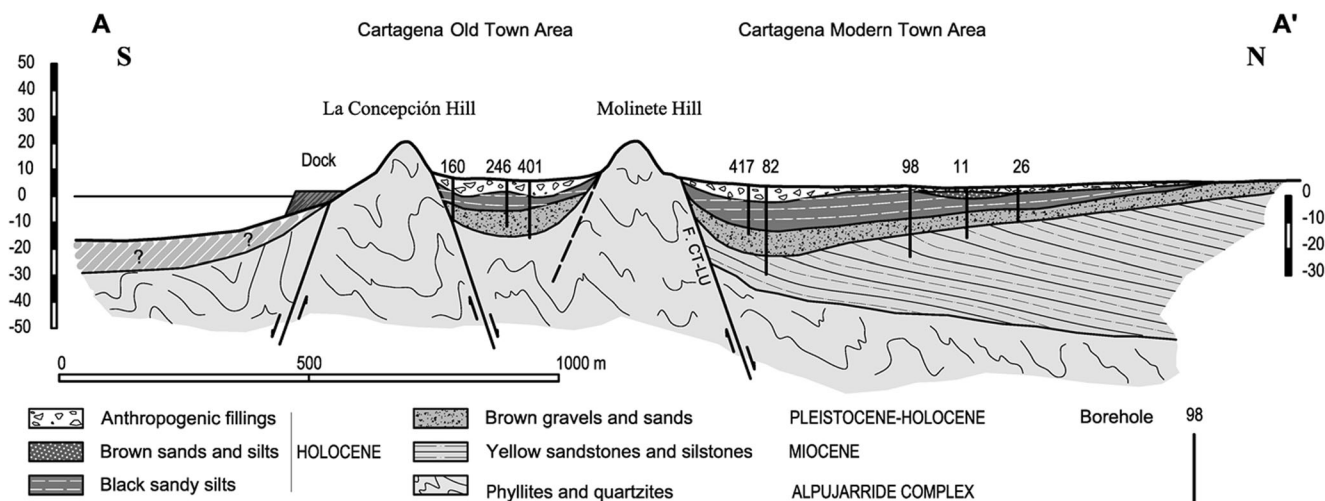


Fig. 4 S-N geological profile of Cartagena area showing the morphology of El Almarjal lacustrine deposits (current study, Arqueotopos project) (the location of the profile can be seen in Fig. 2)

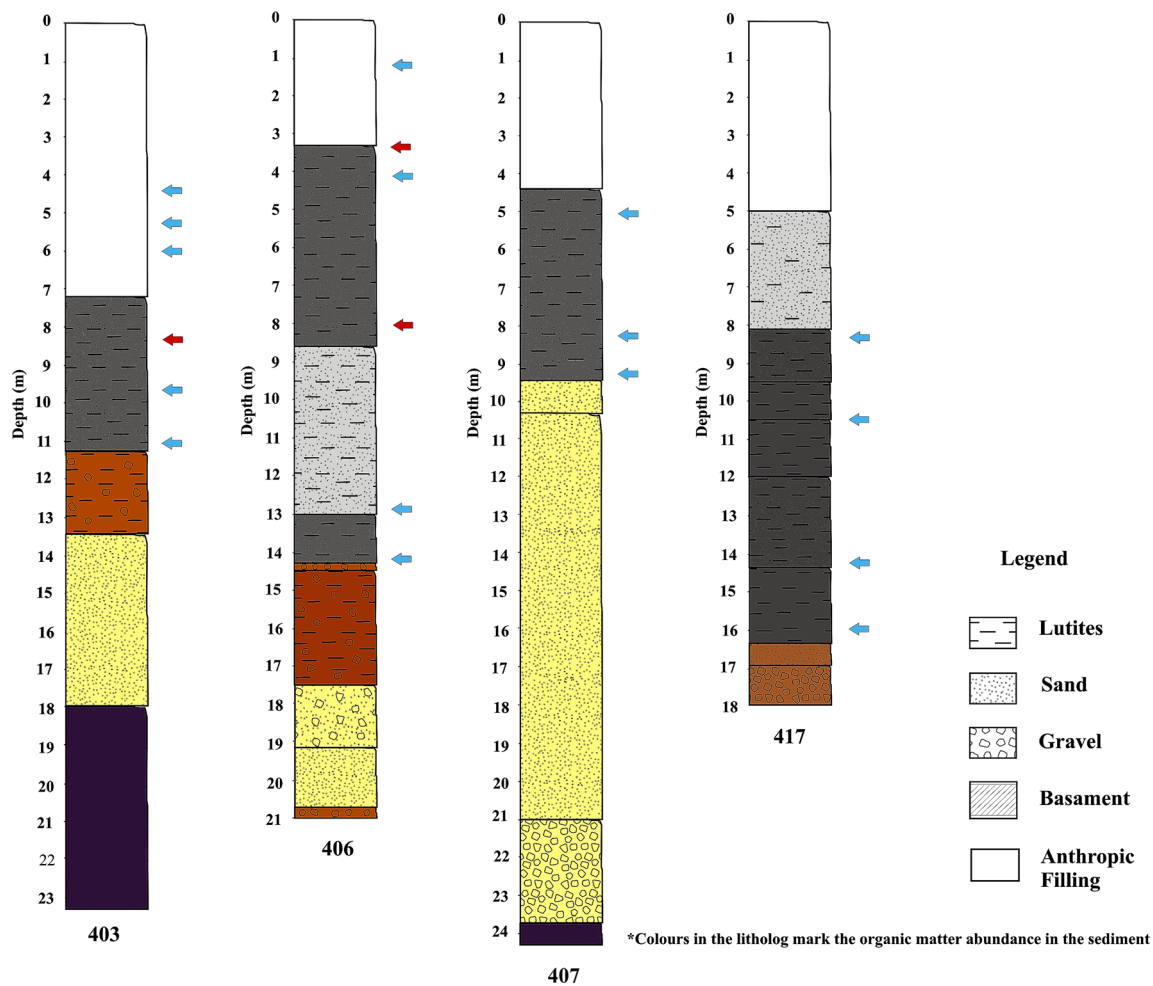


Fig. 5 Lithostratigraphy of boreholes 403, 406, 407 and 417. Grey colours in the lithology marks the organic matter abundance in the sediment. The ^{14}C dating locations in depth are stated to the right of

each core; accepted datings are pointed out in blue, and the discrepant ones are in red (current study, Arqueotopos project)

must be explained as a side effect of sample characteristics (bulk organic matter), being lead enriched horizons a time marker by themselves. The high organic matter concentration precluded also oxidative processes allowing metal mobilisation. In fact, delicate organic components (insect tests, fish scales, seeds) are well preserved.

It is clear that the El Almarjal basin was far from being the depth lake or the sea bottom. On the contrary, it rather was an area with some micro-relief made of very low topographical contrast network of inlets and plant-fixed islands, but it seems that the erosion, transportation and resedimentation did not play a very important role in the final sedimentary architecture. Bioturbation is different because there is a clear presence of burrowing organisms as a saline/brackish water cockle (*C. glaucum*) that burrows some centimetres deep into the soft unconsolidated mud. A more important fact is the presence of plant roots that penetrate some decimetres into the sediments, thus being responsible of a contribution of modern carbon into older sediments.

Based on the ^{14}C datings, the former lagoon of El Almarjal started developing more than 8000 years ago (the earliest records found are dated at 8450 cal years BP) at the same time of the recorded evidences of the Holocene transgressive effect in the Mediterranean (Carmona et al. 2016). Two older values around 13,000 cal years BP have been found in continental gravel samples beneath lagoon facies.

The age of the basal sediments of the Almarjal presents significant differences, depending on the thickness of the sediments and the unequal depth of the basin. In the deepest zones, the basal sediments present ages around 8000 cal years BP, as in the southern section near the peninsula relief (boreholes 403, 406, 407, 417). However, in areas with smaller sediment thickness, their age is lower, as in the northern zone (boreholes 410, 413, 416), where the age of the basal sediments is around 4000 cal years BP. This suggests the influence of successive transgressive effects during the Holocene (Fig. 7).

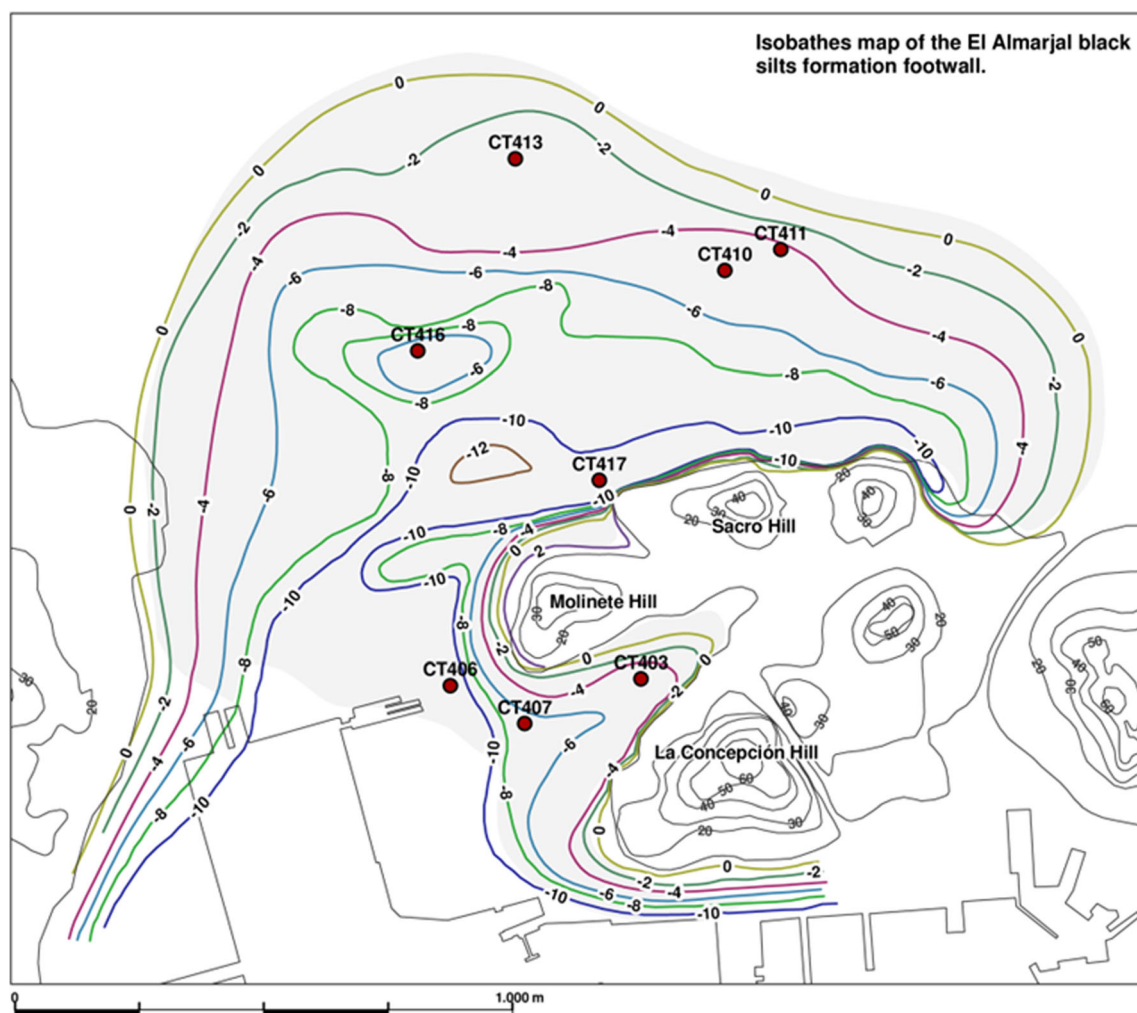


Fig. 6 Isobaths map of the sapropelic silt formation footwall of El Almarjal (only the modern boreholes we have drilled and sampled are represented, but a large number of former geotechnical holes have also been used to compile this map) (current study, Arqueotopos project)

Moreover, in the boreholes next to the old city, the sedimentary record was interrupted in Roman period due to a

significant anthropic infilling of the lagoon southern edge, whereas no infilling was done in northern areas of the basin

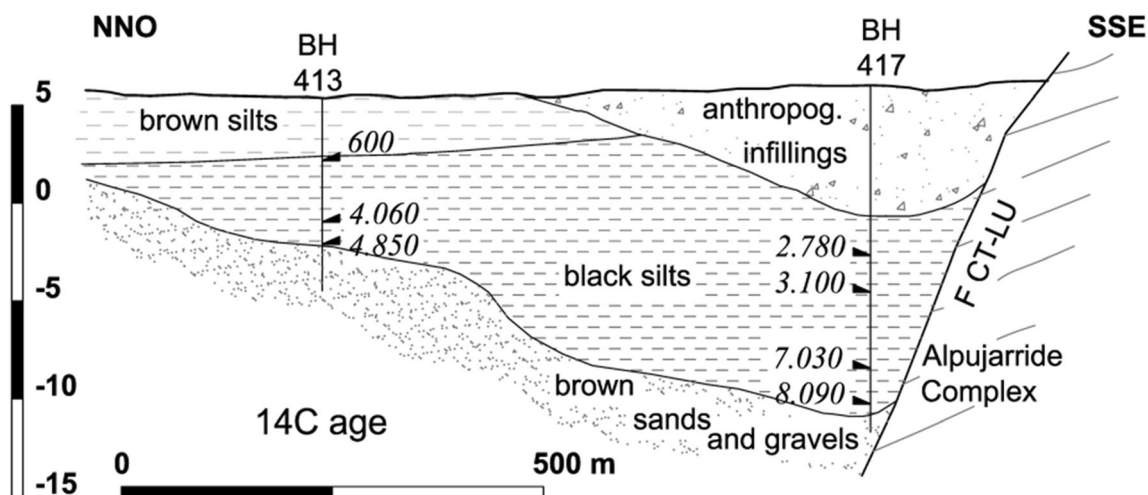
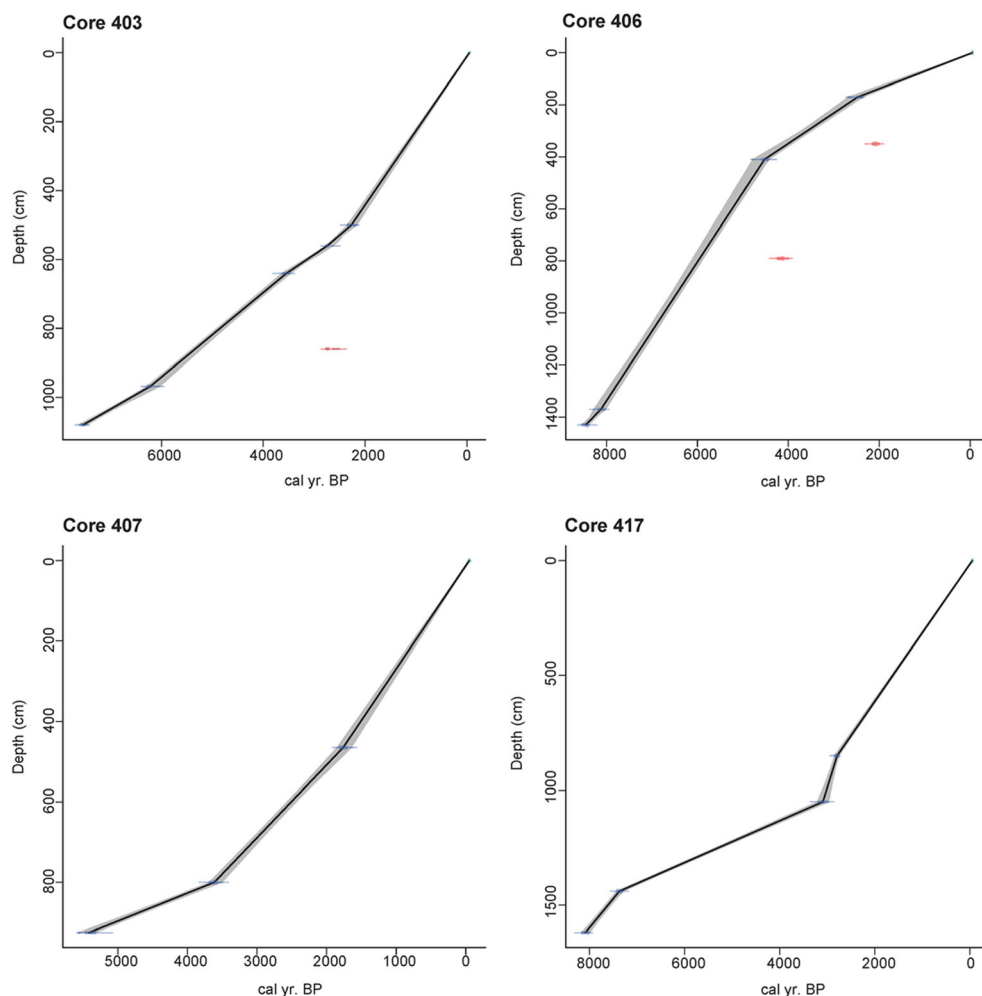


Fig. 7 Geological profile of the Almarjal basin showing the differences of sedimentary records between northern and southern zones. Values along boreholes are the ^{14}C dating (cal years BP) (current study, Arqueotopos project)

Fig. 8 Age models of 403, 406, 407 and 417 borehole cores. Discrepant datings are represented in *red* colour (current study, Arqueotopos project)



and the sedimentary record of the boreholes reaches their more recent ages.

According to the 32 ^{14}C datings obtained (Table I in Annex) and to the result of the age-deep models performed, several anomalous values have been detected, mainly in the 406, 410 and 416 cores, which could explain the inconsistent

points of the corresponding age-depth model graphs, reflected in an older age than the samples below (Fig. 8). As an example, Fig. 9 represents the stratigraphy of the borehole 406, together with the vertical variation of Pb content and the ^{14}C sample dating. This figure shows anomalous age values for two of these samples. In the case of anomalous dates at the top

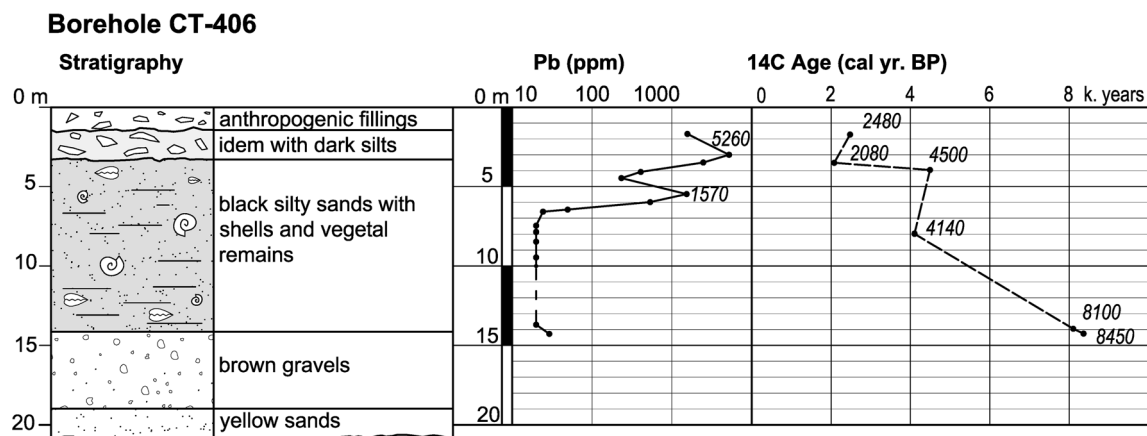


Fig. 9 Stratigraphy, lead records and ^{14}C dating of borehole 406 with two anomalous datings (2480 cal years BP in anthropic level and 4140 cal years BP in the black silt level) (current study, Arqueotopos project)

of the columns, in facies of anthropic origin, it is very likely that as suggested by the material archaeological context, these datings were caused by anthropogenic removals. As for the rest of the discrepant datings, their origin could be in post-depositional sediment reworking phenomena related mainly to bioturbation episodes. To minimise this issue, we have considered the possibility of selecting only those boreholes with reliable datations (407, 413 and 417) and the 403 and 406 except for their two outliers.

Relative to the geochemical composition

The mean values of the samples from El Almarjal show relatively significant metal contents, especially in the case of lead, 692 ppm of Pb, 103 ppm of Zn and 65 ppm of Cu. All these values stand out comparing to other basins mentioned in the bibliographic references, except for the estuary of the Tinto River in Huelva, where these values are even higher (Leblanc et al. 2000). We should also highlight the prominent predominance of lead over zinc despite the latter metal is more abundant at the mineral deposits of Sierra de Cartagena (Manteca and y Ovejero 1992) and that its geochemical mobility is higher. Table 1 shows the elemental statistics of the most significant elements from all the samples.

Borehole metal content varies depending on their horizontal location at the basin (Fig. 10). The Pb, Cu and Zn contents of boreholes closest to the old city (CT403, CT406, CT407 and CT417) are greatly higher than the contents of the farthest boreholes (CT410, CT411, CT413 and CT416). For example, in the case of the period between 2500 and 1700 cal years BP, the mean lead content of the four closest boreholes is 1463 ppm, whereas for the four farthest boreholes is only 267 ppm, roughly a fifth part. This steep contamination

gradient, inversely to the distance to the city, shows that the main contamination source would be inside the city, probably on the area closest to the harbour premises, and undoubtedly caused by the activity of the metallurgical workshops. This same zonality occurs with copper and zinc.

It is also important to mention that the location of the boreholes with the highest contents of heavy metals seems closely related to the two main drainage axes linked to the geomorphology of the old city, conditioned in turn by the fault system, a ENE-WSW riverbed running between the hills of El Molinete and La Concepción and opening to the west to the former harbour area, as well as a NNW-SSE riverbed, almost perpendicular to the former, that runs between the Molinete and Sacro Hills and discharges into the southern swampy area. As a support to the pollution being of urban origin, we should refer to the findings of metallurgical furnace structures from the Roman Republican and Empire eras at several zones of the old town, which are Montanaro street (Madrid Balanza 2005), Beatas-San Cristóbal La Corta streets (Rodríguez García and Hernández Ortega 2008), the Amphitheatre (Pérez-Ballester and Rodríguez Fernández-Trejo 1999), La Merced square-Duque street (Fernandez-Henarejos Jiménez et al. 2005) and PERI CA-4 (Madrid Balanza 2005). To this fact, we must add the great presence of lead ingots inside the vessels found in the harbour and the coast close to it, which undoubtedly came from local industries and the harbour storage houses of the city.

Regarding the metal vertical variability, the analysis of cores shows that the content of some elements, especially Pb, Cu and Zn, varies significantly depending on the depth of the sample and thus on its age. However, other elements, such as Mn, Fe, Sr and Ba, vary more slightly or even remain constant, as it happens with Ni. Regarding the Pb, Cu and Zn

Table 1 Elemental statistics from El Almarjal 92 borehole samples

Element	Mean percent	Standard deviation	Minimum percent	Maximum percent
C	5.1974	2.7128	1730	14.510
Na	0.6879	0.3033	0.245	1570
Mg	1.5769	0.3548	0.771	3196
Al	6.0046	2.0350	2257	9608
Si	16.6840	4.7555	4786	27.640
S	0.8317	0.6190	0.0537	3747
Mn	0.0395	0.0202	0.0084	0.0973
Fe	2.6168	0.9109	0.8334	4491
Ni	0.0032	0.0012	0.0000	0.0057
Cu	0.0065	0.0079	0.0021	0.0645
Zn	0.0103	0.0056	0.0026	0.0320
Sr	0.0595	0.0324	0.0110	0.1991
Ba	0.0314	0.0136	0.0000	0.0699
Pb	0.0692	0.1234	0.0020	0.7093



Fig. 10 Lead zonation at the El Almarjal basin boreholes in relation to their distance to the former peninsula and the old city (period 2500–1700 years BP) (current study, Arqueotopos project)

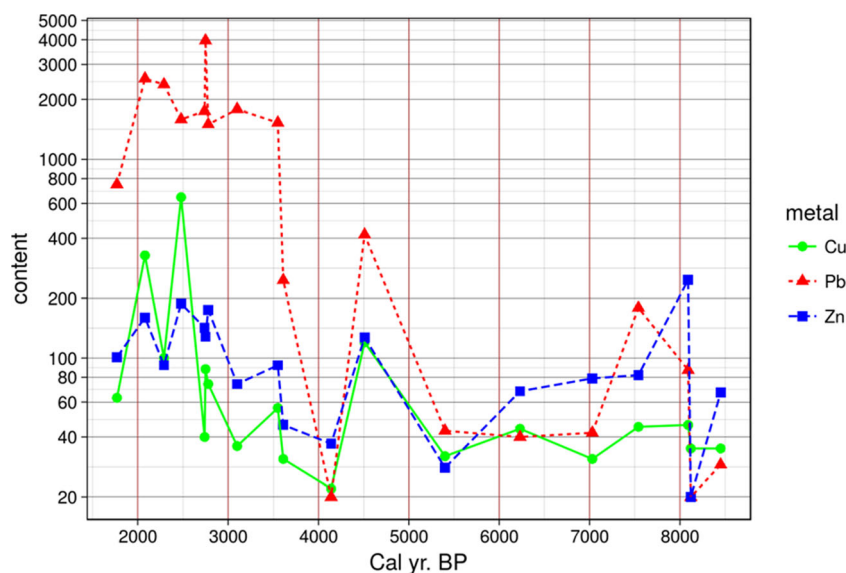
groups, the contents are low at the lower section of the boreholes, approximately below the height of -3 m bsl (which may vary from -2 to -5), but on the top section above that height, the values progressively increase. This variation is especially significant in the case of lead, where the values exponentially vary from one sample to another.

For the purposes of finding the model of vertical variation in heavy metal contents, the graphics of metal contents versus ^{14}C age of the most representative boreholes have been created. As the thickness of the black silt level changes laterally for the different holes, then correlation is based on the sample age dating. In all of them, there is a low content in heavy metals in the lower part of the columns in samples prior to 5000 cal years BP. The first meaningful growth of these metals is recorded in the 406 core at the 4500 cal years BP, increasing mostly from the 3500 cal years BP. The highest contents are concentrated between the 2000 and 3000 cal years BP with a maximum near the 2800 cal years BP. The graphic models of content metals/age correlation

for each borehole are shown as supplementary information (Figs. I and II in Annex).

On the other hand, given the horizontal zonation mentioned above, the model of vertical variation of metals has also been studied for the two differentiated zones, which are the area of the immediate holes to the city and the area next to the most distant holes. Both models are significantly different from each other. The first model includes boreholes in the southern area of the lagoon, closest to the old city (or proximity area) (403, 406, 407 and 417), whereas the second model includes boreholes in the northern region, farther from the city (or distal area) (410, 413 and 416). Figures 11 and 12 include the corresponding graphics. The graphic relating to the set of 19 samples from the proximity area (Fig. 11) shows the following general features: low metal content below 5000 cal years BP, a significant peak at 4500 cal years BP and a great content above 3600 cal years BP, which remains in a plateau until 2000 cal years BP and then starts to decrease. The graphic relating to the 11 samples of the distal area (Fig. 12) is

Fig. 11 Age and heavy metal correlational graphic of the boreholes closest to the old city (403, 406, 407 and 417) (current study, Arqueotopos project)



noticeably different. It shows a low metal content at the start of record (4850 cal years BP), which rapidly increases and reaches a first peak sometime before 4000 cal years BP. Later on, it reaches the maximum value around 3000 cal years BP and then starts to decrease, falling to values below 200 ppm of Pb after 1500 cal years BP. However, this graphic is highly undetermined, as there are only three samples above 3000 years BP. More specifically, there are no samples available from the interval between 3000 and 2000 cal years BP, a period without doubt very important in mining. Consequently, we consider that this graphic has little significance.

After analysing the two sets, we consider that to obtain the representative picture of the geochemical evolution at this basin, we must group together both sample sets.

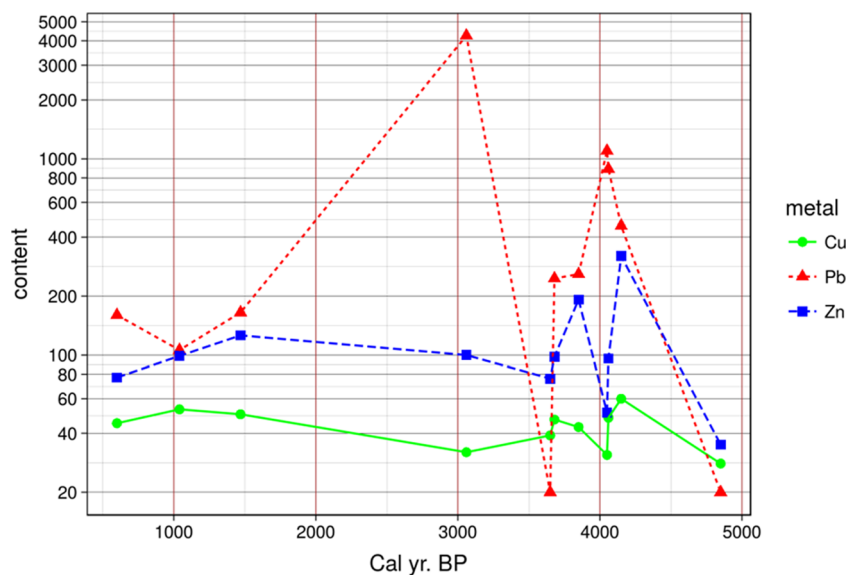
For a first analysis of the geochemical evolution of the basin, we have grouped all the dated cores (30 samples from the seven studied boreholes). Figure 13a shows the evolution

of metal content versus the ^{14}C age of the samples, whereas Fig. 13b shows the said evolution depending on the depth of the samples. Both graphs should be similar in theory, but in reality, they present strong differences. These differences may be due to the presence of anomalous samples, in relation with post-depositional sediment reworking phenomena, previously cited, which may eventually produce a potential bias in the model.

For this reason, in a second analysis, we have focused on the samples where there is no evidence of alteration, excluding the samples with discrepant values. With this criterion, 19 of the 30 available samples were selected, 403 cores (except 403.4); 406 cores (except 406.1 and 406.4); and 407, 413 and 417 cores. Figure 14 corresponds to the resulting model.

The exclusion of samples considered discrepant has slightly improved the correlation coefficient age dating/sample depth, which goes from 0.84 to 0.88. Therefore, we consider

Fig. 12 Age and heavy metal correlational graphic of the boreholes farthest to the old city (410, 413 and 416) (current study, Arqueotopos project)



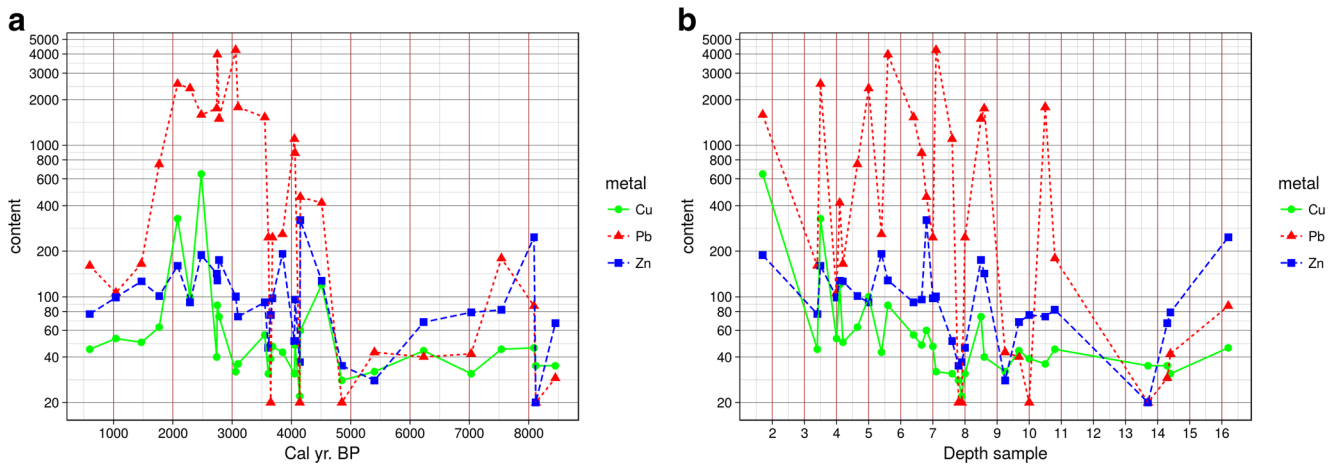


Fig. 13 Correlational graphics of the whole core set. **a** Metal contents versus sample age datings. **b** Metal contents versus sample depth (current study, Arqueotopos project)

that this last model is more representative of the evolution of the metal content of the basin.

Evolution through time of the metal contents at the El Almarjal basin

According to the model of Fig. 14, the metal contents in samples older than 4500 cal years BP are systematically low, with mean values considered as the local background of the respective elements, which are 40 ppm of Pb, 37 ppm of Cu and 54 ppm of Zn (in order to estimate the lead background, we have excluded a value of 179 ppm, whereas we have excluded another value of 247 ppm in the case of zinc, as we considered both values to be anomalous).

At 4500 cal years BP, we find a significant increase of metals, but especially in lead, which from that time stands out in several magnitude degrees from the other two metals. We believe that this may be the beginning of anthropic contamination due to mining and metallurgic activities based on lead. From this age on, we observe a period where low and

high values of these metals alternate, presumably because of mining and metallurgical intermittent activities on the area. In the case of lead, this period lasts until 1700 cal years BP. The highest contents of lead are at the period between 3550 and 1700 cal years BP, with a mean value upon 2000 ppm. Regarding the copper and zinc, the increases are comparatively unimportant, generally not exceeding 200 ppm, and are concentrated especially in the interval between 2000 and 2600 cal years BP. Lastly, metal contents decrease strongly between 1700 and 600 cal years BP.

A very interesting fact about this content distribution through time is the relation between the lead and zinc values. The contents of zinc are always significantly lower than lead contents in those samples after 4500 cal years BP. However, zinc values are clearly higher during the previous period (zinc background 54 ppm, lead background 40 ppm). This behaviour is consistent with the idea that during the period before 4500 cal years BP, the metal contribution to the basin would only come from natural origins. The content of Zn is thus higher than the Pb content according with its greater

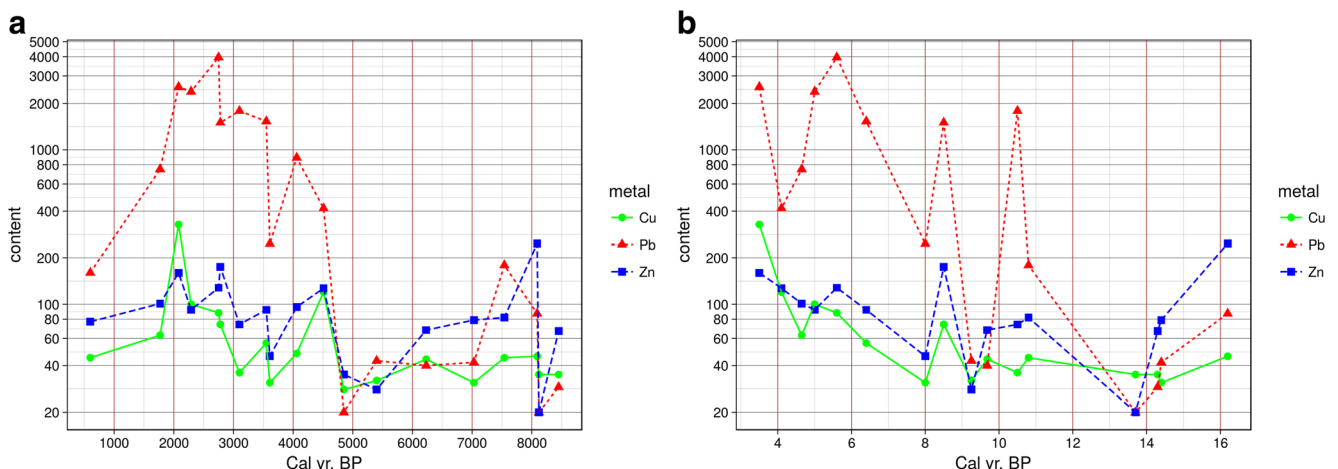


Fig. 14 Correlational graphics of the core set excluding samples with discrepant values (current study, Arqueotopos project)

Table 2 Variances retained by each components

	Dimension 1	Dimension 2	Dimension 3	Dimension 4	Dimension 5
Variance	1.903	1.252	0.744	0.684	0.417
Percent of variance	38.058	25.034	14.886	13.681	8.341
Cumulative percent of variance	38.058	63.092	77.978	91.659	100.000

abundance in the source area (Manteca y Ovejero 1992). Nevertheless, starting on that date, the lead content significantly surpasses that of zinc because of the birth and development of an early mining and metallurgical industry based in copper and lead ores with zinc playing only an accessory role.

Regarding the principal component analysis carried out on the individual heavy metals, depth and age variables, a Kaiser, Meyer and Olkin (KMO) factor adequacy of 0.53 has been obtained, which shows that performing a PCA analysis is adequate (Table III in Annex). The following Tables 2 and 3 show the results of variance percentages from analysis and the correlation matrix of the chosen components. The first two dimensions of principal components retain a 63.092% of the variability.

The PCA showed a grouping of different parameters (Fig. 15). Two main components (PC) have been identified. They account for 63.092% of the total variance, with the 38.058% being explained by the dimension 1 (or component 1), where Cu and Pb appear to be strongly grouped by this component. Moreover, this component seems to be linked to the age and depth of the samples, suggesting that it is an anthropogenic factor (mining-metallurgical activity). The graphic biplot of the individuals and variables in the principal component analysis is included in Fig. III in Annex.

On the other hand, the dimension 2 (or component 2) come determined by the Al variable and explains the 25.034% of cumulative variance that could suggest a governing natural factor. The Zn is correlated with both factors (0.61 and 0.56). Note that dimension 1 is positively related with Cu, Pb and Zn and negatively with age and depth variables. This would prove that high metal values are directly related to the youngest values of the age variable and also with the lower depth.

Table 3 Correlation matrix between variables and dimensions

Variables	Dimension 1	Dimension 2	Dimension 3
Depth sample	−0.736	0.213	0.194
Cu	0.771	−0.187	−0.419
Pb	0.597	−0.343	0.716
Zn	0.611	0.560	0.021
Al	0.190	0.860	0.133
Supplementary continuous variable			
Cal years BP	−0.609	0.175	−0.048

Discriminating natural and anthropogenic contamination: the enrichment factor

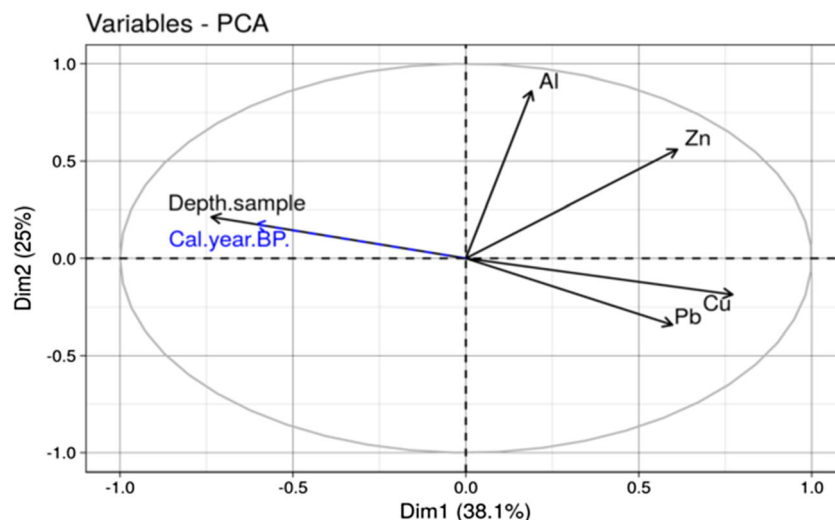
In environmental studies, it is important to discriminate between natural and anthropogenic sources of the pollution. Geochemistry bibliography offers various methods for assessing anthropogenic influence. In some situations, the lead isotopic signature method is used, based on the Pb^{206}/Pb^{207} relation, especially in cases of superposition of current and ancient contamination. In our case, we have chosen for the current phase of the study the enrichment factor (EF) method, frequently used in metal contamination studies of sedimentary basins, referenced by Reimann and de Caritat (2005), Galuska and Migaszewski (2012) or Delgado et al. (2012), amongst others.

To apply this method, the knowledge of the typical element concentration in a specific area and in a specific medium (local background) is essential. It is also necessary to choose a reference element of terrigenous character (normalizing element), not affected significantly by the anthropic process like aluminium (Schütz and Rhan 1982; Shotyk 1996). In the EF method, the ratio between the metal concentration and the normalizing element value of every sample, $([M]/[N])_{sample}$, is compared with the similar ratio from the local background, $([M]/[N])_{background}$. The formula for the case of lead is as follows: $Pb\ EF = ([Pb]/[Al])_{sample}/([Pb]/[Al])_{background}$.

The EF values close to 1 mean that the metal content is caused by natural processes (natural contamination), whereas when $EF > 1$, we need to assume as well the existence of anthropogenic input of metals apart from natural contamination (Chatterjee et al. 2007; Huang et al. 2009; Delgado et al. 2012). Other authors propose a stricter criterion and believe that the contamination is basically anthropogenic when the EF factors are higher than 10 (Reimann and de Caritat 2005; Galuszka and Migaszewski 2011).

In the case of El Almarjal basin, we have studied the evolution through time of the EF for the selected three metals in the sedimentary records, which are lead, copper and zinc. The graphic for lead is presented at a scale that is lower than the other elements because of its high values and its great dispersion. We have used the EF 10 value as a reference to differentiate the periods with undoubtedly predominant anthropic lead contamination. For copper and zinc graphics, we have adopted the EF2 value to differentiate the anthropic contamination episodes.

Fig. 15 Graphic projection of the variables in the principal component analyses



On the other hand, to verify the representativeness and validity of the evolution model of EFs, we have developed this one from two databases: (a) totality of the dated samples and (b) database excluding samples with anomalous dating. In the first case, the number of samples is 30 (Fig. 16) and in the second 19 (Fig. 17). The comparison of both graphs allows verifying that the general characteristics of the metal EF evolution models

are very similar in the two cases. Common characteristics to highlight are as follows:

The first sign in these graphics of a clear anthropic metal origin, presumably related to the mining and metallurgical activities, can be found at 4500 cal years BP, with a Pb EF of 13.1, a Cu EF of 4.8 and a Zn EF of 3.24. In stages prior to that age, EFs present systematically low values, with the exception of an anomalous Zn EF of 4.49, located at

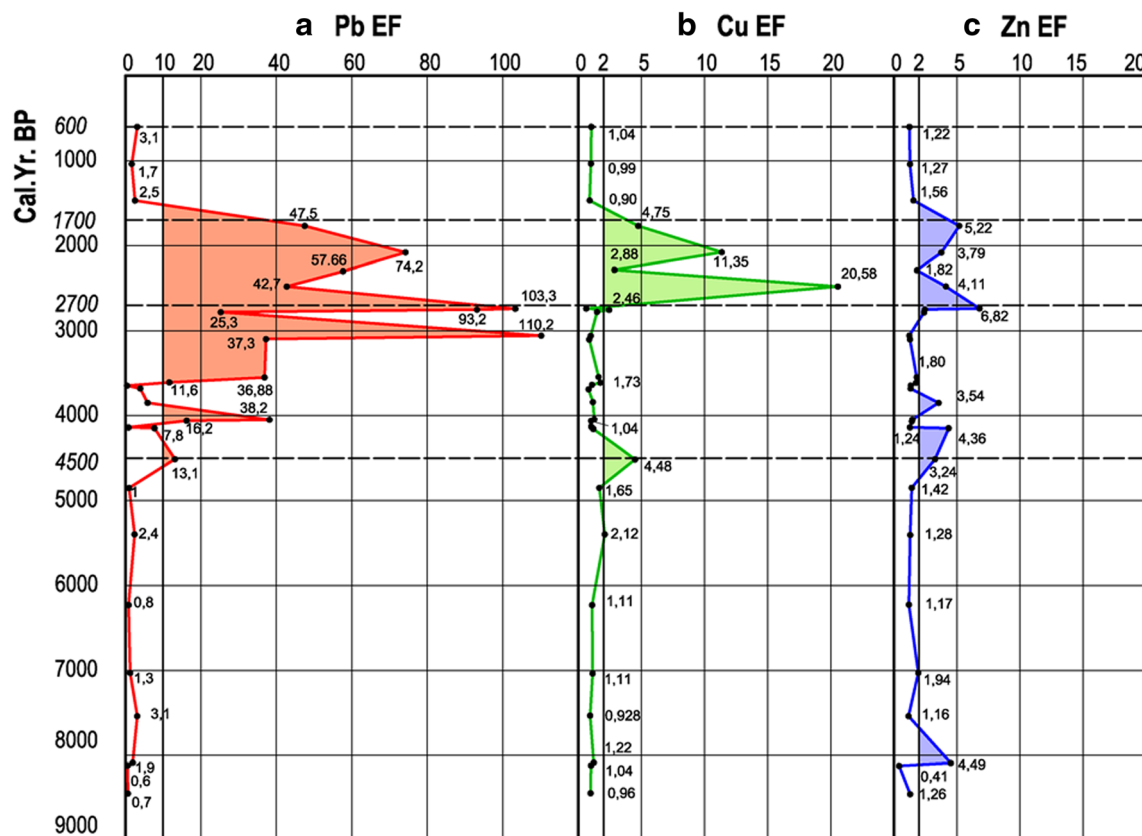


Fig. 16 Evolution through time of the EF for lead, copper and zinc in the lacustrine sediments from El Almarjal (whole core set; current study, Arqueotopos project)

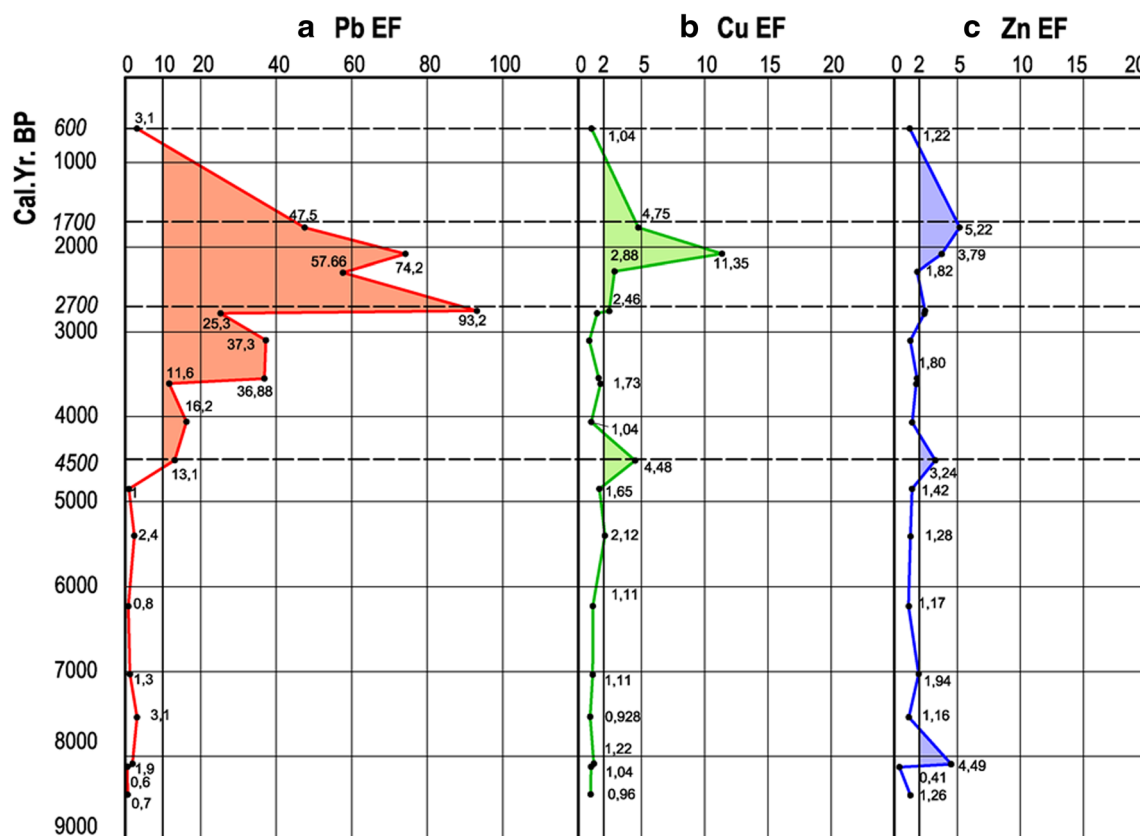


Fig. 17 Evolution through time of the EF for lead, copper and zinc in the lacustrine sediments from El Almarjal (depurated core set; current study, Arqueotopos project)

8.090 cal years BP (anomaly already mentioned in “[Results and discussion](#)” section, in probable relation to a fire episode).(Carrión et al. 2003; Carrión et al. 2010)

In the case of lead, the most important and sustained contamination stage starts from 3550 cal years BP and is maintained up until 1700 cal years BP, reaching EF values higher than 90. It is noteworthy that the highest lead contamination peaks took place long before the Roman Empire, between the Late Bronze Age and the Iron Age.

Cu and Zn contamination is clearly less important and more limited in time, presenting certain differences between both graphs. According to Fig. 15a, the main stage of contamination of both metals took place from 2700 until after 1700 cal years BP, whilst according to Fig. 15, its extension is somewhat lower, from 2300 until after 1700 cal years BP.

After 1700 cal years BP, the anthropic metal contamination decreases in both graphs, meaning a lower mining and metallurgic activity in the old city area.

A comparison between the two graphs shows that although the number of samples used in each case is very different, the EF evolution models obtained are very similar, mostly the dating of the beginnings of anthropic contamination and the delimitation of the main contamination stage. We can infer that it is an indicator of the consistency and representativeness of these models.

Comparison with the basins of the South-West of the Iberian Peninsula

The lead contamination evidences found at the El Almarjal basin on a date as early as 4500 cal years BP (Late Chalcolithic) in addition to the significance of that contamination have led us to contrast our data with those from the studies carried out on the South-West of the Iberian Peninsula at the Iberian Pyrite Belt. We can consider this area as a relatively analogous environment to that of Cartagena, considering the presence of polymetal deposits and a millenarian mining history, and thus, we would expect to obtain similar results.

At the estuary of the Tinto River in the province of Huelva, on the basis of the geochemical study of a 15-m-deep borehole, it has been determined the existence of a first level of anthropic contamination with values of 2500 ppm of Pb, 400 ppm of Cu, 240 ppm of Zn and 900 ppm of As. The estimated age for this level is 4500 cal years BP (Leblanc et al. 2000; Davis et al. 2000; Olías and Nieto 2015), which corresponds to the beginning of the mining and metallurgic activities at the Iberian Pyrite Belt deposits. The importance of the metal contamination in this case is clearly higher than at El Almarjal in Cartagena.

Moreover, at the estuary of the Guadiana River, on the border between Spain and Portugal, the sedimentary record of the last 13,000 years based on two boreholes of 51 and 63 m of depth has been studied, respectively (Delgado et al. 2012), and the existence of anthropic metal contamination (Pb, Co, Ni, Mn, Zn and Cu) starting 4500 years ago has been also checked. However, the contamination rate in this case is clearly lower than the one registered at El Almarjal in Cartagena. The maximum registered values of Pb, Zn and Cu are 41.3, 87.0 and 54.0 ppm, respectively.

In summary, we can say upon the present study, and according to the studies mentioned above, that the age of the beginning of metal anthropic contamination in the El Almarjal basin is very similar to the corresponding age in the estuaries of the Guadiana and Tinto Rivers; then, we may infer that the mining and metallurgic activities started during the same period at both mining districts.

Conclusions

The swamp-lagoon sediments from El Almarjal are affected by a noteworthy heavy metal contamination, especially from lead, with mean values of 692 ppm of Pb, 65 ppm of Cu and 103 ppm of Zn.

The metal contents of the boreholes show a strong zonality inversely proportional to their distance to the old city. This fact suggests that the main contamination source would be at the city itself in relation to the metallurgical activities. Besides, the boreholes showing the highest contamination are in the area next to the mouth of the two thalwegs of the former peninsula where the city stood, which may prove that there is a link between a great part of the contamination and the runoffs of the old city.

The evolution through time of the metal contents and its EF allows us to get to conclusions about the chronology and origin of the metal contributions to the basin, as explained below.

Before 4500 cal years BP, the sample contents are systematically low and their mean values (40 ppm Pb, 37 ppm Cu and 54 ppm Zn) are considered as the respective backgrounds for those metals. The corresponding enrichment factors are close to the value of 1, being consistent with natural contributions associated with the erosive and sedimentary cycle. There is an exception for the zinc at 8090 cal years BP with an anomalous 4.49 EF that may be due to various processes not investigated yet.

The first significant metal increase takes place at 4500 cal years BP with a Pb EF of 13.1, a Cu EF of 4.48 and a Zn EF of 3.24. These enrichment factors point out that origin of the contamination is anthropic, caused by the beginning of a primitive mining and metallurgical industry based in copper and lead mixed ores. There is a second and short increase in anthropic lead contamination at 4050 years, with an EF of 16.2.

The most important and lasting period of anthropic contamination referring to lead took place from 3550 cal years BP until the end of the Roman Empire at 1700 cal years BP. During this period, according to Fig. 16, an average Pb EF of 62 is reached. If based on Fig. 17, the average BP EF is lower, about 48. The highest EFs are found specifically between the Late Bronze Age and the Iron Age. It is noteworthy that the highest lead contamination peaks happened long before the Roman Empire. On the other hand, the main anthropic contamination inputs for copper and zinc take place during the Roman period from 2200 to 1700 cal years BP, with a maximum Cu EF of 11.36 at 2200 cal years BP and a maximum Zn EF of 5.22 at 1750 cal years BP.

The anthropic contamination of the basin decreases after 1700 cal years BP as shown by the corresponding EFs, with values close to 1 for Cu and Zn and close to 2 for Pb. These values prove that there was a strong decrease metallurgical activity on the environment of the old city. The contamination by zinc, a metal unknown in the old times, must be considered as a secondary consequence of the mining and metallurgical activities focused in lead and copper due to the close relation between these three metals at the polymetal deposits of Cartagena district.

The high lead content is an evidence of the great environmental contamination at an already existing possible primitive settlement standing on the site occupied by the future city of Carthagonova, caused by mining and metallurgical activities throughout different periods, which must have negatively affected the health of the inhabitants of the city. The high lead content in the bone rests found at archaeological excavations in the city from periods when the metallurgical activity was strong (Martínez-García et al. 2005) seems to confirm this fact.

Lastly, we must highlight that according with this study, the origin of anthropic metal contamination in the area of Cartagena due to the beginning of mining is contemporary (4500 cal years BP) to that of the basins located on the South-West of the Iberian Peninsula at the Iberian Pyrite Belt area (Leblanc et al. 2000; Davis et al. 2000; Delgado et al. 2012; Olías and Nieto 2015).

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