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A century of sediment metal contamination of Mar Menor, Europe's largest saltwater lagoon

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ABSTRACT

Coastal enclosed ecosystems, such as lagoons, are vulnerable to anthropogenic impacts because they favor the accumulation of contaminants from the surrounding watersheds, particularly in their sediments. Europe's largest saltwater lagoon, the Mar Menor (SE, Iberian Peninsula), is a highly impacted ecosystem and the first in the continent to be granted personhood rights. Based on a high-resolution spatial and temporal dataset, we present the historical reconstruction of metal contamination in this ecosystem during the last century. Our results highlight that sediment metal contamination has been mainly driven by the development of the mining industry in the nearby Sierra Minera de Cartagena-La Unión in the late 19th and until the mid-20th century when premeditated mining spills were forbidden. Runoff from former mining areas still transported metals to the lagoon even after mining ceased in the 1990s. The southern sector of the lagoon, closest to mining-affected ephemeral streams, is the most impacted by metal contamination and holds the highest metal stocks. Stocks since 1900 for the entire lagoon reached values of 9200, 1.6, 450, 270, 10,000, and 12 tons of Pb, Hg, As, Cu, Zn, and Ag, respectively.

Maxima concentrations were reached in the mid-20th century, with values of 3400, 0.53, 100, 50, 3700, and 5.5 mg·kg^{-1} for Pb, Hg, As, Cu, Zn, and Ag. Afterward, while some metals' concentrations declined, others were still supplied to the sediments through runoff from former mining areas and sources related to urban expansion. Metal concentrations reported in this study surpass sediment quality guidelines and are generally higher than those found in similar ecosystems globally. Current surface metal concentrations are lower than during most of the 20th century, and sediments seem to retain metal stocks effectively. However, climate change and eutrophication could increase the risk of metal remobilization, further impacting this already vulnerable ecosystem. This study emphasizes the need for attention to managing coastal enclosed ecosystems, where global change impacts can exacerbate the impact of legacy contamination.

1. Introduction

Metals, derived from natural (e.g., geological formations) or human-related sources (e.g., mining or fossil fuel combustion), reach marine coastal environments through different pathways. These include atmospheric deposition (Guerzoni et al., 1999; Sánchez Bisquert et al., 2017),

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fluvial inputs (Omwene et al., 2018; Zhang, 1999), submarine groundwater discharge (Alorda-Kleinglass et al., 2019; Trezzi et al., 2016), and sewage effluents (Chen, 2022; Papakostidis et al., 1975). Trace amounts of certain metals can serve as nutrients for primary producers, but they can be toxic at elevated concentrations (e.g., Zn, Mn, Ni, and Cu)(Jordi et al., 2012), while others are only hazardous to biota (e.g., As or Hg). The anthropogenic inputs of these elements to the environment have increased significantly over the last decades (Morelli et al., 2012; Palanques et al., 1998). In this context, As, Cd, Cr, Cu, Ni, Pb, V, and Zn have been recognized as Potentially Toxic Elements (PTEs) (Pourret and Hursthouse, 2019), which can persist in aquatic ecosystems (Dang et al., 2021). In addition to their concentrations, the toxicity of these compounds depends on their speciation and thus bio-availability (Bayen, 2012), as well as the organism affected (Langston, 1990). For humans, concerns primarily center around the potential for bioaccumulation up the food chain and its associated health risks (Sharifuzzaman et al., 2016).

Metals are particulate-bound elements that can be sorbed, flocculated, and coprecipitated (Karlsson et al., 1987; Virtasalo et al., 2023). Transport, accumulation, and resuspension of sediment particles and associated metals in marine coastal systems are controlled by dynamic processes, mainly currents and waves (Nittrouer and Wright, 1994). The low energy of these processes in semi-enclosed ecosystems such as coastal lagoons, along with their morphology, limits the water exchange with the ocean, favoring the accumulation of fine sediments and their associated contaminants (Kjerfve, 1994; Frignani et al., 1997). Coastal lagoons are characterized by high biological productivity (Alongi, 2020) and are crucial in providing valuable ecosystem services such as provision of food (Newton et al., 2018). They occupy 13 % of coastal areas worldwide (Kjerfve, 1994), where 30 % of the global population lives (Kummu et al., 2016). Therefore, these ecosystems are impacted by anthropogenic activities, often leading to eutrophication or metal pollution, which adversely affect water and sediment quality, as well as biota health (Ferreira et al., 2023; Huseen and Mohammed, 2019; Newton et al., 2014; Pignotti et al., 2018).

Metal contamination can be particularly relevant in lagoons hydraulically connected to current and former mining sites (Fernandez et al., 2006). This is the case of Mar Menor, located in the southeast of the Iberian Peninsula, the largest saltwater lagoon in Europe. This ecosystem is included in the Ramsar Convention for Wetlands and the European Natura 2000 network, in addition to containing several regionally protected landscapes. Its climatic conditions and natural resources have encouraged the development of economic activities that menace the ecosystem's equilibrium, namely coastal urban development and intensive agricultural practices, added to past mining exploitation (Conesa and Jiménez-Cárceles, 2007). These activities have affected important resources for local livelihoods like fisheries, and highly valued ecosystem services such as tourism and landscape (Perez-Ruzafa et al., 1991; Velasco et al., 2018).

Several studies have reported large metal contamination in sediments of the Mar Menor coastal lagoon (García and Muñoz-Vera, 2015; Marín-Guirao et al., 2005b; Pacheco, 2010; Pérez-Ruzafa et al., 2023; Serrano et al., 2019), with concentrations as high as 5700 mg·kg^{-1} for Pb, up to three times higher than in other areas greatly affected by metal pollution, such as the industrial site of Venice lagoon (Bellucci et al., 2002). The high concentrations reported in Mar Menor have been linked to the former mining activities in the district of Sierra Minera de Cartagena-La Unión, located a few kilometers to the southwest of the lagoon, which started in Carthaginians' time and reactivated at an industrial level in the 19th century (Vilar Ramírez and Egea Bruno, 1985), until ceasing in 1991. Other infrastructures that have been pointed as sources of metals to Mar Menor are nautical activities related to the eight yacht ports located on its shore due to the release of Cu contained in antifouling paints (Jones and Bolam, 2007; Pérez-Ruzafa et al., 2023).

Most of the previous studies on sediment metal contamination in the lagoon have solely focused on surface samples (Marín-Guirao et al., 2005b; Pacheco, 2010; Serrano et al., 2019), while those that have used dated sediment cores lacked high spatial resolution across the lagoon (García and Muñoz-Vera, 2015; Pérez-Ruzafa et al., 2023). To better understand the drivers of metal fluxes into the sediments and their temporal evolution, historical reconstructions of metal contamination inferred from multiple dated sediment cores can help identify the processes influencing metal accumulation in sediments, such as sedimentation dynamics, land use changes like urbanization (Garcia-Orellana et al., 2011), and policy changes like environmental regulation (Palanques et al., 2017), in addition to tracing contaminant sources and transport pathways (Heim and Schwarzbauer, 2013; Valette-Silver, 1993).

This study aims to examine the spatial distribution and decadal variability of sedimentary metal concentrations, fluxes, and sources over time in Mar Menor by using ^{210}Pb dated sediment cores evenly distributed across the lagoon. In this study, results will focus on Pb, Hg, As, Cu, Zn, and Ag due to their historical importance, different potential sources, and ecotoxicological and health impacts (Harada, 1978; Pérez-Ruzafa et al., 2023). This approach will help to comprehend one of the causes of the environmental stress this location has endured in the past century, how it relates to socioeconomic changes, and inform future management plans.

2. Methods

2.1. Study site

Mar Menor is a hypersaline coastal lagoon located on the Iberian Peninsula Mediterranean coast (Fig. 1). It spans over 135 km^2, with average and maximum depths of 4.5 and 6.5 m, respectively (Baudron et al., 2015). It has only one surface watercourse that maintains a regular flow of fresh water, the Albujón, resulting from a water transfer that started in 1979 and a subsequent irrigation-induced phreatic rising (García-Pintado et al., 2007). The lagoon is connected to the Mediterranean Sea by three main channels within the 22 km-long sandy bar known as La Manga (Perez-Ruzafa et al., 1991). Water circulation in the lagoon is mainly wind-driven (Pérez-Ruzafa et al., 2005), with a primary circulation gyre in the center and two other circulation subsystems north of El Estacio channel and south of the Barón and Perdiguera Islands (Fig. 1). Most of the lagoon sediment is currently muddy and enriched in organic matter with percentages close to 25 % in central surface sediments (Belando et al., 2021), and anoxic conditions at depth, while sandy sediments can be found in the shallowest areas and close to Las Encañizadas inlet (Pérez-Ruzafa et al., 2023).

Based on circulation patterns and characteristics, the lagoon can be divided into three sections: the north (north of the main outlet to the sea, El Estacio channel), the center (main circulation gyre, anticlockwise), and the south (south of the islands, with downward currents from the Albujón and El Estacio circulating to the southernmost part of the lagoon) (Fig. 1). The northern sector receives little runoff from ephemeral streams, and it is influenced by the natural connection to the sea (Las Encañizadas). The central sector is mainly affected by the El Estacio channel and the discharge of the permanent watercourse, the Albujón. Finally, the southern sector of the lagoon has historically

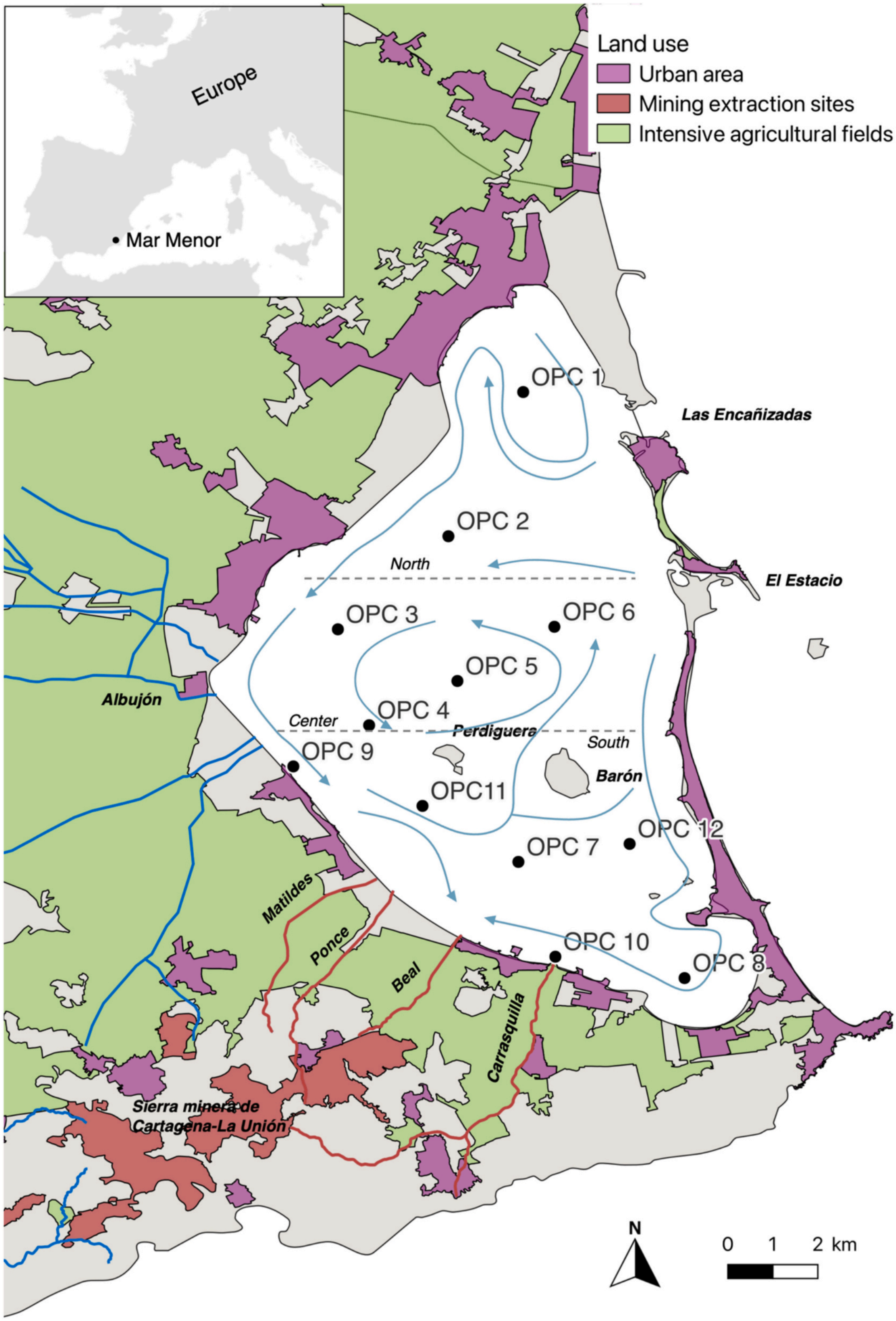

Fig. 1. Location of the sampled sediment cores and surrounding land uses based on the CORINE land cover (2018) in the Mar Menor coastal lagoon, SE Iberian Peninsula. Red lines represent watercourses that run through former mining areas. The dashed grey line in the lagoon separates what is considered as north, center, and south in this article. The blue arrows show the present day lagoon circulation, based on Pérez-Ruzafa et al. (2023), and J. Gilabert (personal communication, June 20, 2024). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

received mining tailings' debris through ephemeral streams that carry water only after significant rainfall events. In this work, the north-center-south division has been used to present and discuss our data.

The influence of human activities in the lagoon has lasted for more than three millennia (Perez-Ruzafa et al., 1991). The mines from the Sierra Minera de Cartagena-La Unión started being exploited 3500 years ago by the Carthaginians and the Romans (Conesa and Jiménez-Cárceles, 2007; Oen et al., 1975; Robles-Arenas et al., 2006), mainly to obtain silver and lead from argentiferous galena (Manteca Martínez, 1992). Four ephemeral streams connect the mining area to the southern portion of Mar Menor, allowing mining debris to reach the lagoon (Cuevas et al., 2023). Mining activities started to affect the local environment in the mid-19th century when the modern mining industry resurfaced through smallholder exploitations and was followed by industrialized activities in the Sierra Minera (Sánchez i Bassols et al., 2008). By the end of the century, the mining district had already become a national force as a lead producer (Vilar Ramírez and Egea Bruno, 1985). Production was always centered on lead and silver, but zinc and iron were also extracted (Manteca Martínez, 1992) with remarkably high productivity until the mid-20th century (Cerdà i Domènech, 2020; Conesa and Jiménez-Cárceles, 2007).

As a consequence of a strong social mobilization (Pérez-Ruzafa et al., 1987), from 1955 onwards the authorities banned mining-derived spills in the lagoon, and ponds were constructed to store sludges (Vilar Ramírez et al., 1991). While these measures were implemented, open-pit mining started, majorly based on the exploitation of lead and zinc (García and Muñoz-Vera, 2015). This increased the number of processed materials (Conesa and Jiménez-Cárceles, 2007). In 1968, most of the subterranean mining activities ended (García and Muñoz-Vera, 2015), and in 1991 all mining ceased (Marín-Guirao et al., 2007). However, mining tailings continued to be eroded and metals reached the lagoon, particularly during typical Mediterranean torrential rains (María-Cervantes et al., 2009; Marín-Guirao et al., 2005a). In 2022, dams were installed to stop the transport of these wastes to the lagoon (Ministerio de Transición Ecológica y Reto Demográfico, 2021, 2023).

Beyond mining activities, other important impacts in this ecosystem include intensive agriculture since the 1990s and a far-reaching urban expansion fueled by the tourism industry since the 1970s (Fig. 1), which have caused major ecological changes such as the introduction of allochthonous species and turning the ecosystem progressively more eutrophic, with biota mass mortality events happening recurrently since 2016 (García-Pintado et al., 2007; Instituto Español de Oceanografía, 2020). In 2022, after a strong social mobilization, a Popular Legislative Initiative (ILP) triggered the creation of Law 19/2022, which recognizes the lagoon's right to exist as an ecosystem and to evolve naturally, as well as its right to protection, conservation, maintenance, and restoration (Fuchs, 2023), being the first European ecosystem to be acknowledged as such (Krämer, 2023).

2.2. Sampling and sample processing

Twelve sediment cores were collected in December 2020 and June 2021 (Fig. 1). Sampling sites for cores OPC9 and OPC10 were accessed from the beach by foot, while the deeper cores were collected by divers. All cores were extracted by manual hammering using clear PVC tubes (120 cm long, 8.5 cm internal diameter). Cores have been grouped into three regions: north (OPC1 and OPC2), center (OPC3, OPC4, OPC5, OPC6 and OPC9) and south (OPC7, OPC8, OPC10, OPC11 and OPC12).

Cores were sliced every 1 cm and samples were stored in plastic bags at 4 °C. Representative subsamples were dried at 65 °C until constant weight was achieved. They were then ground with a mortar and pestle except for subsamples used for grain size analysis. Dried subsamples were kept in sealed plastic bags at room temperature for ^{210}Pb and metal concentration analysis.

Dry bulk density (g·cm^{-3}) of each slice was calculated by dividing dry weight per sample volume. In this study, we display sediment profiles using accumulated mass (g·cm^{-2}) instead of depth (in cm), since the former is not affected by compaction processes (Gifford and Roderick, 2003; Wendt and Hauser, 2013).

2.3. Analytical and dating methods

Grain size fraction along each sediment core was determined using a Horiba Partica LA-950v2 particle-size analyzer, with an accuracy of 0.6 % and precision of 0.1 %. The contents of gravel (>2 mm, including mollusk shells), sand (2 mm–63 μm), silt (63–4 μm) and clay (<4 μm) was obtained for each sample.

The specific activities of ^{210}Pb along each sediment core (Fig. A1) were determined by measuring its decay product ^{210}Po in radioactive equilibrium. Known amounts of ^{209}Po were added to sediment aliquots. Aliquots were digested with a microwave oven via the addition of HNO_3 and HF through a temperature ramp that reached 180 °C for 10 min, plus neutralization of fluorides with H_3BO_3 following Sanchez-Cabeza et al. (1998). Po isotopes were autoplated onto silver disks and their emissions were measured by alpha spectrometry on Passivated Implanted Planar Silicon (PIPS) detectors (CANBERRA, Mod. PD-450.18 A.M). The concentrations of ^{210}Pb in the samples were calculated through decay—ingrowth corrections from sampling to counting, and accounting for blanks and detector backgrounds. The activity of ^{210}Pb supported by the presence of ^{226}Ra in the sediment matrix was determined in selected samples by gamma spectrometry. These samples had been stored in sealed containers for 3 weeks to attain equilibrium between ^{226}Ra and its short-lived decay products. Gamma measurements were performed in a high-purity Ge well-type detector (CANBERRA) and ^{226}Ra was determined through the 352 keV emission line of ^{214}Pb. Sediments were dated with two ^{210}Pb dating models: Constant Rate of Supply (CRS) (Appleby and Oldfield, 1978), and Constant Flux: Constant Supply (CF:CS) (Krishnaswamy et al., 1971). For each core, the most appropriate model was selected by comparing the results from both models and validating them (Arias-Ortiz et al., 2018; Smith, 2001). Sediment mass accumulation rates (MAR) (g·cm^{-2}·yr^{-1}) for the datable period were obtained for each complete core based on the chosen model, using the excess ^{210}Pb horizon as a guide.

For the determination of bulk metal concentrations (mg·kg^{-1}), the same digestion as for ^{210}Pb was followed, plus filtering through 0.45 μm diameter nylon filters. The analyses were carried out using an Inductively Coupled Plasma Mass Spectrometer (ICP-MS; Agilent 7900 model). Analyzed metals comprise Na, Al, Si, Ca, Ti, Cr, Fe, Ni, Cu, Zn, As, Sr, Ag, Cd, and Pb. Standard reference materials NIST 2702 and IAEA 457 were analyzed for metal concentration recovery factors, which can be found in Table 1. Reported results are not corrected.

Total Hg in sediment samples was determined by a thermal decomposition step, followed by amalgamation and atomic absorption spectrophotometric detection according to the U.S. EPA method 7473 (U. S. EPA, 1998), and using AMA-254 (Altec s.r.l., Czech Republic). The accuracy of measurements was repeatedly checked by analyzing lake sediment reference material MESS-3 (National Water Research Institute, Canada). The measured values were in the certified range for all analyses. Precision determined by replicate analysis was better than 5 %.

Table 1
Certificate and measured concentrations for standard reference materials and recovery factors. Concentrations are shown in mg·kg^{-1} or %, the latter indicated by *).

Metal (mg·kg^{-1} or %*)	NIST 2702			IAEA457		
	Reference	Measured average ($n = 3$)	Recovery factor (%)	Reference	Measured average (n = 3)	Recovery factor (%)
Ag	–	–	–	1.9 ± 0.4	2.1 ± 0.1	110
Al	8.4 ± 0.2*	4.5 ± 1.3*	54	83,000 ± 3000	45,000 ± 12,000	54
As	45 ± 2	40.5 ± 0.1	90	10 ± 1	10.5 ± 0.1	110
Cd	0.817 ± 0.011	1.3 ± 0.2	160	1.09 ± 0.08	1.2 ± 0.1	110
Cr	350 ± 20	290 ± 10	83	144 ± 8	120 ± 1	83
Cu	–	–	–	370 ± 20	350 ± 4	95
Fe	7.9 ± 0.2*	6.4 ± 0.3*	81	41,000 ± 2000	38,000 ± 1100	93
Ni	75.4 ± 1.5	60.6 ± 2.7	80	53 ± 3	48 ± 1	91
Pb	132.8 ± 1.1	130 ± 2	98	105 ± 7	110 ± 2	100
Sr	120 ± 3	48 ± 17	40	137 ± 10	50 ± 20	36
Ti	0.8267 ± 0.006	0.7 ± 0.1	85	–	–	–
Zn	485 ± 4	350 ± 13	72	430 ± 30	340 ± 8	79

2.4. Metal stocks, accumulation rates, and enrichment factors

Metal stocks (g·m^{-2}) were calculated by integrating metal concentrations in sediments down to a common age horizon (approximately 1900) in dated cores that reached this date (all but OPC9 and OPC10). Metal quantities (g) were added up until the sediment section that corresponded to 1900 (uncertainties included) and then normalized by area. Metal concentrations between analyzed sections were interpolated linearly when a result was missing or nonreliable. The total stock values (tons) for the lagoon were estimated by multiplying the median stock value by the area of the lagoon. Metal accumulation rates (g·m^{-2}·yr^{-1}) were calculated as the product of metal concentration in a sediment layer and the mean MAR for that core.

Enrichment factors (EF; Eq. (1)) were calculated to estimate the relative level of contamination shown by the metal concentrations in the sediment cores (Martinez et al., 2023). EFs are used to normalize concentrations of metals (Me) concerning a common element in the continental crust (Ti in this study). Ti has been used over Al and Fe due to the low recovery factors obtained for Al (Table 1), and because Fe is a product of the mining activities conducted in the region and therefore there are substantial contributions of this element in the area, rendering it unsuitable as a normalizing element (Boës et al., 2011; Sakan et al., 2015). EFs are calculated as follows:

$$EF = \frac{\left[\frac{Me}{Ti}\right] sample}{\left[\frac{Me}{Ti}\right] background} \quad (1)$$

where $[Me/Ti]_{sample}$ is the normalized concentration of a metal in a sample, and $[Me/Ti]_{background}$ is the normalized concentration of a metal in the background samples. The background samples' concentrations were considered as the metal concentrations for the deepest layers of each core, which date before the 19th century. The calculated results represent a conservative estimate since the maximum depth of the collected cores (75 cm) might still be affected by smallholder mining occurring at the time.

2.5. Interpolation and statistical analysis

Inverse Distance Weighting (IDW) was used to explore the spatial distribution of MARs, metal stocks, and metal concentrations within the study area (software QGIS, default power value of 2). Spearman correlation analysis was conducted to evaluate significant relationships between metal concentrations, dry bulk density, and grain size fractions. Normality for metal distribution in each lagoon core and region (north, center, south) was checked for with a Shapiro-Wilk test. Kruskal-Wallis' rank sum test was used to detect if metal differences by core or sector were significant. Dunn test with Bonferroni correction was performed for pairwise comparisons among cores and sectors and reported p-values were adjusted to this correction. The suitability of the data for factor analysis was checked with the Kaiser-Meyer-Olkin criterion (Kaiser, 1974). Major sources of metals were then identified by a Principal Component Analysis (PCA) that included metal concentrations, grain size, and dry bulk density variables. The R package *rshift* (Room et al., 2023) was used to detect significant temporal shifts in Pb, As, Hg, Cu, Zn, and Ag sediment concentrations with depth (i.e., time). STARS algorithm (Sequential t-test Analysis of Regime Shifts) (Rodionov, 2004) was used for cases with a normal distribution (significance level = 0.05, cut-off length = 3 cm, which can amount to 2 to 15 years). The resulting Regime Shift Index (RSI) quantifies the size of the shift (the more positive, the greater). Lanzante's non-parametric L-Method (Lanzante, 1996) was performed for non-normal distributions (significance level = 0.05). Its resulting p-values also express the size of the shift.

3. Results

3.1. Sediment grain size and dry bulk densities

The dominant grain size fraction in most of the samples was silt, with an average value of 65 % and a maximum content of 89 % (Fig. C1), except for samples collected in shallow areas in front of the mouth of ephemeral streams (cores OPC9 and OPC10), which had a greater sand content. This was specifically the case for the first 20 cm of OPC9 (average sand proportion of 82 %) and the entire OPC10 (average value of 96 %). The second most significant fraction was either clay or sand. Clay was the second most prevalent fraction in the surface layers of OPC4 and OPC11, the middle sections of OPC5, and all sections of OPC6 and OPC7. Sand was the second most common fraction in some deep sections of OPC4 and OPC11, surface sections from OPC5, and all sections of OPC12. Cores in the north and center-west (OPC1, OPC2, and OPC3) exhibited an increase in sand content from mid-depths (~15 cm) downcore. The upper sections of these north-western cores presented a high silt content (from 44 to 85 %).

Generally, dry bulk density (DBD) (Fig. C2) increased with depth

Table 2
Sediment mass accumulation sediment rates (MAR in g·cm^{-2}·yr^{-1}) for the dated cores in the lagoon.

Region	Core	MAR (g·cm^{-2}·yr^{-})
North	OPC1	0.045 ± 0.002
	OPC2	0.073 ± 0.018
Center	OPC3	0.054 ± 0.016
	OPC4	0.13 ± 0.02
	OPC5	0.074 ± 0.013
	OPC6	0.052 ± 0.007
South	OPC7	0.15 ± 0.03
	OPC8	0.09 ± 0.04
	OPC11	0.120 ± 0.007
	OPC12	0.106 ± 0.008

across most sediment cores, from 0.19 g·cm^{-3} to 3.2 g·cm^{-3}. Cores located nearshore close to stream mouths (OPC9 and OPC10) had greater DBD fluctuations along the sediment profile and generally presented higher values, on average (1.6, as opposed to 0.7 g·cm^{-3} for the rest).

3.2. Lead-210 dating

Lead-210 specific activities showed decreasing trends punctuated with deviations from the general pattern until reaching constant values at depths from 8 to 26 cm (approx. Accumulated mass of 5 to 18 g·cm^{-2}). Average ^{226}Ra specific activities were 24 ± 4 Bq·kg^{-1} ($n = 2$), 29 ± 10 Bq·kg^{-1} ($n = 6$), and 12 ± 5 Bq·kg^{-1} ($n = 7$) in the northern, central, and southern sectors of the lagoon, respectively (Fig. A1). Some mixing was present along the entire cores OPC2 and OPC7, as inferred by the ^{210}Pb profiles. Cores located in front of ephemeral streams' mouths (OPC9 and OPC10) showed no excess ^{210}Pb specific activities, thus preventing the determination of age models. Since these cores are sand-rich and ^{210}Pb is not scavenged by sand it cannot be discarded that there has been sand accumulation over time. The ^{210}Pb-specific activity profile for core OPC4, located close to the Albujón mouth (Fig. 1), showed depleted activities in the upper sediment sections, which were also characterized by higher dry bulk density values and higher sand content (Figs. C1 and C2).

Mean MARs derived from ^{210}Pb are depicted in Table 1. They averaged 0.059 ± 0.011, 0.060 ± 0.005, and 0.120 ± 0.017 g·cm^{-2}·yr^{-1} in the north, center, and south of the lagoon, respectively (Table 2). The highest MAR values are in the south, except for OPC4.

3.3. Depth profiles of metal concentrations and accumulation rates

Concentration profiles of Pb, Hg, As, Cu, Zn, and Ag with depth (expressed as accumulated mass: g·cm^{-2}) and time for representative cores of each sector are presented in Fig. 2. Profiles for the rest of the cores can be found in Section F of the Appendices.

The highest sediment metal concentrations were found in the south, with levels decreasing spatially toward the north. Preindustrial metal concentrations were similar for all metals except Cu in the three sectors. Metal concentrations increased significantly in all the regions from the late 19th century until the 1950s (See Regime Shift Indices and p-values in Table G1). The highest concentrations for all metals except Cu were found in the southern sector during this industrial mining period with values of 3400, 0.53, 100, 50, 3700, and 5.5 mg·kg^{-1} for Pb, Hg, As, Cu, Zn, and Ag.

After the 1950s, concentrations of most metals (except Cu) started to decrease significantly in the center and, particularly, the south of the lagoon. However, some metals followed different patterns. Mercury concentrations in the north of the lagoon rose again significantly in the mid-1970s. In the south, after Hg concentrations increased until the 1950s, they followed a significant decrease during the 1980s. From the 1980s to the 2000s, there were small Hg increases in the north and the center. Arsenic presented significant increases in the central sector during the 1960s and 2000s. For Cu, significant increments were observed in the 1970s in the north and the center. These concentrations stayed similar or increased, especially in the northern sector at the beginning of the 21st century. Ag concentrations increased in the southern sector during the 1980s and in the northern sector in the 2000s.

Metal accumulation rates (Fig. 3) consistently showed higher values in the southern region than in the rest of the lagoon after 1920. Accumulation rates peaked in that sector from the 1930s to the 1950s for Pb (3.0 ± 1.4 g·m^{-2}·yr^{-1}), Hg (0.5 ± 0.2 mg·m^{-2}·yr^{-1}), As (0.07 ± 0.03 g·m^{-2}·yr^{-1}), Zn (3.3 ± 1.5 g·m^{-2}·yr^{-1}) and Ag (5 ± 2 mg·m^{-2}·yr^{-1}). For Cu, the highest value occurred in the 2000s (0.04 ± 0.02 g·m^{-2}·yr^{-1}).

3.4. Spatial distribution of metals

Metal concentrations in sediments are summarized in Table D1. As already observed in the depth profiles, metals like Pb, Hg, As, Cu, Zn, and Ag showed the highest concentrations in the south. In contrast, the highest concentrations of Ca and Sr were found in the north, and the central part of the lagoon presented the highest concentrations of Ti, Al, Ni and Cr. Significant differences ($p < 0.01$) (Table E1) were observed among the lagoon sectors regarding Pb, Hg, As, Cu, Zn, and Ag concentrations, with higher values in the south.

Metal stocks accumulated since the start of the industrial mining period (~1900) were highest in the southern sector (Fig. 4), coincident with higher metal concentrations and greater sediment accumulation rates. The highest values for these stocks were 210 g·m^{-2} for Pb, 0.031 g·m^{-2} for Hg, 5.2 g·m^{-2} for As, 4.6 g·m^{-2} for Cu, 230 g·m^{-2} for Zn and 0.29 g·m^{-2} for Ag. Total stocks for the whole lagoon (135 km^2) accumulated during the last century were estimated to be 9100, 1.6, 450, 270, 10,000, and 12 tons of Pb, Hg, As, Cu, Zn, and Ag, respectively.

3.5. Correlation and Principal Component Analysis (PCA)

The correlation analysis showed that the silt content was positively and significantly correlated with most metals, with higher correlations for Cr, Ti, and Ni ($r > 0.5$) and negatively and significantly correlated with Sr ($r < -0.5$) and Ca ($r > -0.5$) (Table H1). Sand fraction content positively correlated with Sr (r > 0.5), Ca ($r < 0.5$), and negatively correlated with Cr, Ni, and Ti (r < −0.5). Most metals correlated negatively and significantly with DBD since metal content was typically higher in surface sections, which tend to be less dense than their deeper counterparts.

A Principal Component Analysis (PCA) was computed with metal concentrations, dry bulk densities, and grain size data from sections dated from 1900 to 2020. The two principal components explained a cumulative variance of 60 % (Fig. 5). Principal Component 1 (Dim1, horizontal axis), explaining 39 % of the variability, was mainly influenced by Cu, Pb, Zn, Ag, Fe, As, Cd, and Hg with contributions between 6 % and 11 % for each metal. Principal Component 2 (Dim2, vertical axis), explaining 21 % of the variance, was influenced by Sr, Ni, Cr, Ti, Si, sand, and silt (contributions between 6 % and 11 %).

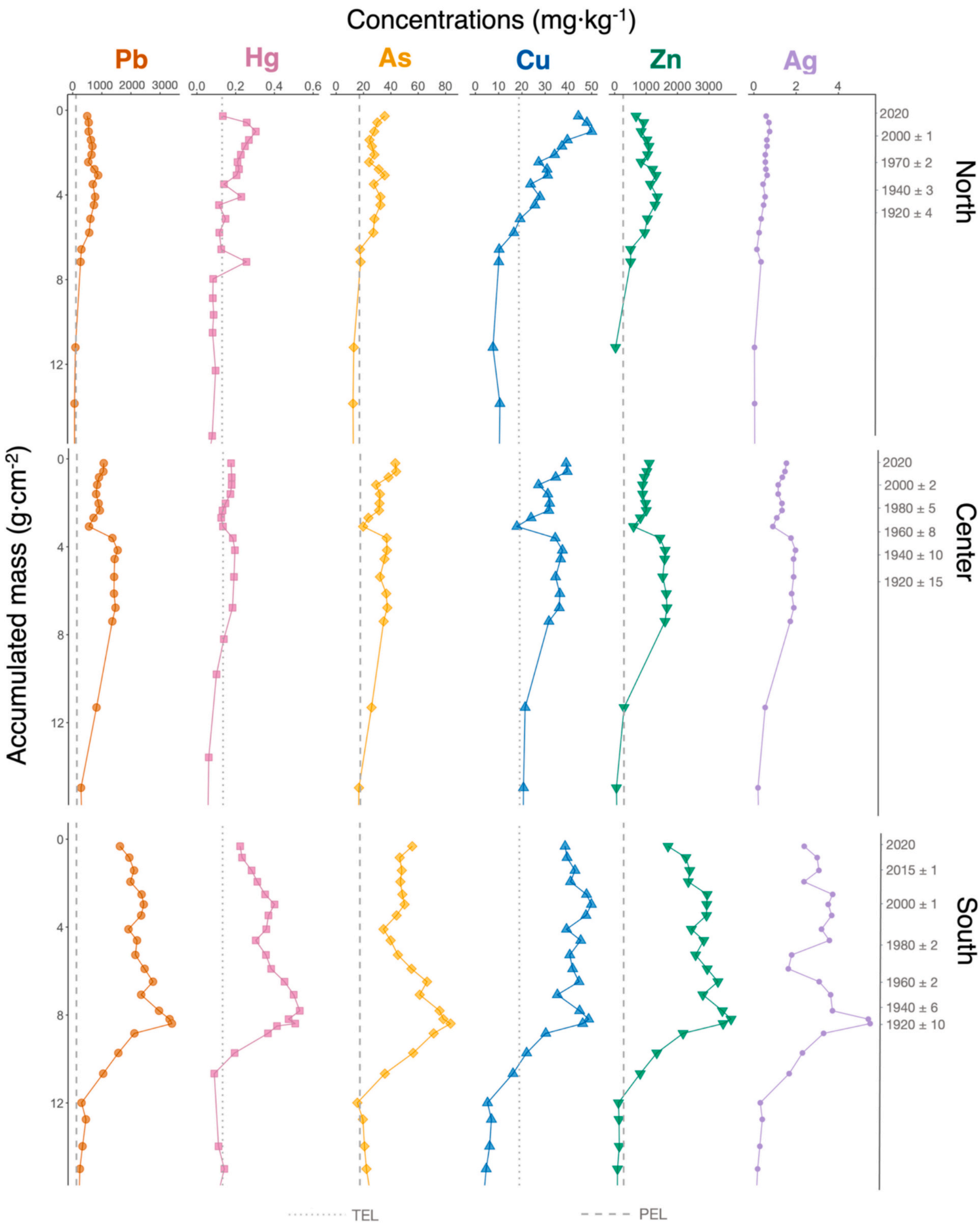

Fig. 2. Concentration profiles of Pb, Hg, As, Cu, Zn, and Ag of selected cores representative of the different lagoon sectors (OPC1 in the north, OPC6 in the center, and OPC8 in the south). Profiles for all the other cores are in Section F of the Appendices. The dotted lines depict the Threshold Effects Levels (TEL) (Hg = 0.13 $mg \cdot kg^{-1}$ and Cu = 18.7 $mg \cdot kg^{-1}$) and the dashed lines the Probable Effects Levels (PEL) (Pb = 112 $mg \cdot kg^{-1}$, As = 17 $mg \cdot kg^{-1}$, and Zn = 271 $mg \cdot kg^{-1}$).

4. Discussion

4.1. Last century sedimentation patterns in Mar Menor are driven by inputs from streams and mining materials

Sedimentation processes in the Mar Menor coastal lagoon result from different sediment delivery mechanisms and redistribution processes. The higher MAR values in the southern sector indicate a combined influence of the proximity of inputs from streams and the secluded southern circulation that promotes sediment accumulation in the southeastern region (Fig. 1). Mining-derived sediments constitute a major fraction of the depositional materials in this region, as evidenced

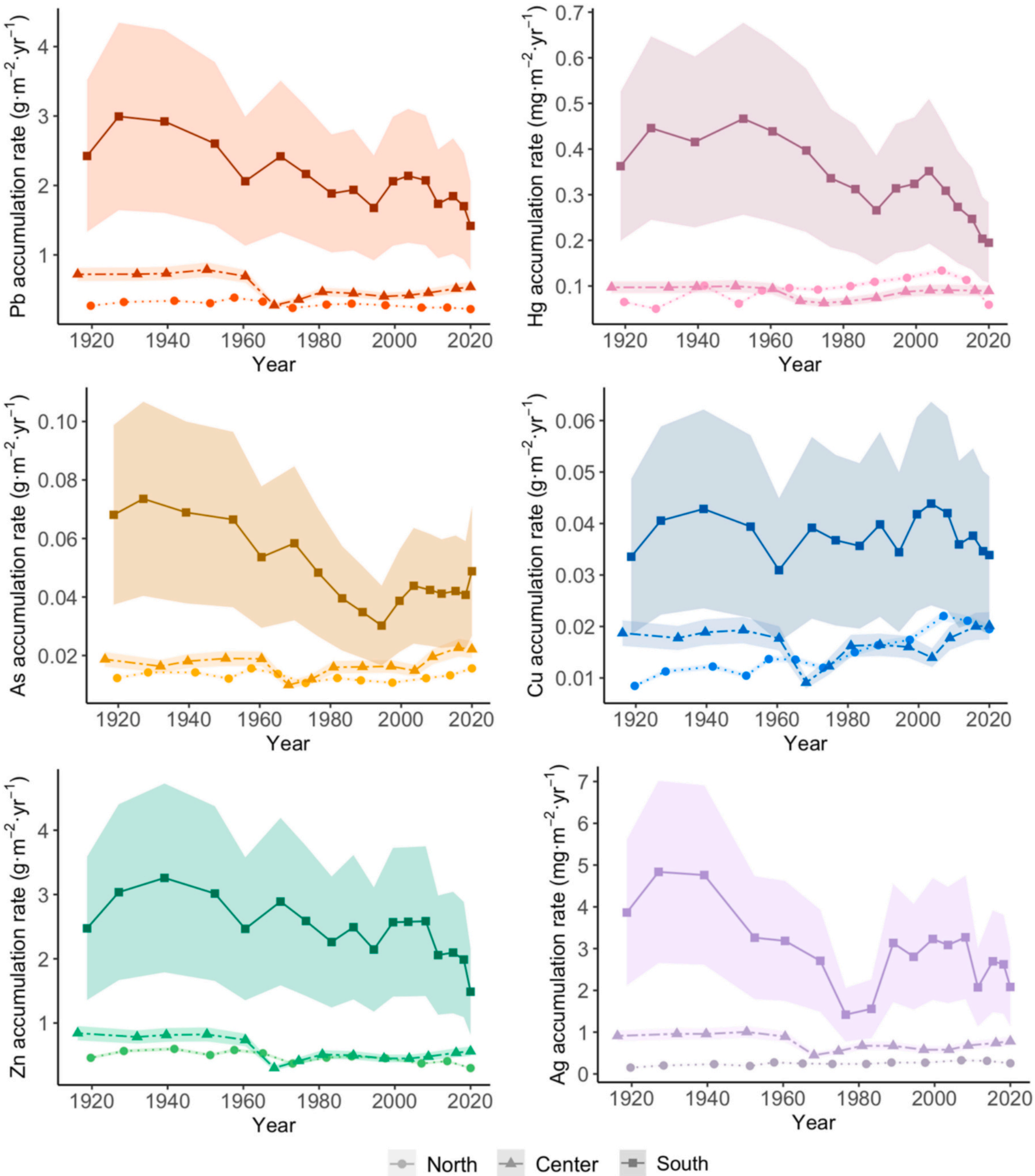

Fig. 3. Pb, Hg, As, Cu, Zn, and Ag accumulation rates during the last century for cores OPC1 (north), OPC6 (center), and OPC8 (south). Note that units are $g \cdot m^{-2} \cdot yr^{-1}$ for Pb, As, Cu, and Zn and $mg \cdot m^{-2} \cdot yr^{-1}$ for Hg and Ag. The shaded area represents the associated error.

by its high metal concentrations and stocks. In the shoreline cores (OPC9 and OPC10), the absence of excess ^{210}Pb is produced by the negligible accumulation of fine sediment fractions locally (Figs. A1 and C1). For core OPC4, located near the shore and the Albujón watercourse, the ^{210}Pb depleted sections might be the result of a recent deposition event (Burnett et al., 2023; Walsh and Nittrouer, 2004) from extreme storms (cut-off lows) which transported large amounts of sediments to the lagoon (Machado Toffolo et al., 2022). The event-corrected dating model indicated that the initial sediment layer beneath the rapid deposit sections dates to 2016, coinciding with the first cut-off low event that had relevant effects on the lagoon mass mortality episodes.

Sandy materials in the lagoon have been associated with marine influences, such as inputs from the sandy barrier that separates the lagoon from the Mediterranean Sea (García and Muñoz-Vera, 2015), and to shoreline sediments in front of watercourses. In contrast, finer sediments, such as clay and silt, have been linked to terrigenous inputs (Pérez-Ruzafa et al., 2023). Metals like Ti and Sr further highlight this terrestrial or marine origin, respectively. Sr and Ca, probably originated from calcareous organisms, presented their highest concentrations in the north (Table D1), suggesting a marine source through seawater inflow via sandbar channels rather than fluvial inputs. High concentrations of metals like Ti or Al but also Cr and Ni in the central sector were attributed to lithogenic sources and the runoff input from the Albujón watercourse (Cuevas et al., 2023). In the southern sector, significantly higher concentrations of Pb, As, Cu, and Ag were linked to the proximity to the mouths of the mining-affected ephemeral streams, which would preferentially deliver these metals, combined with the effect of lagoon circulation.

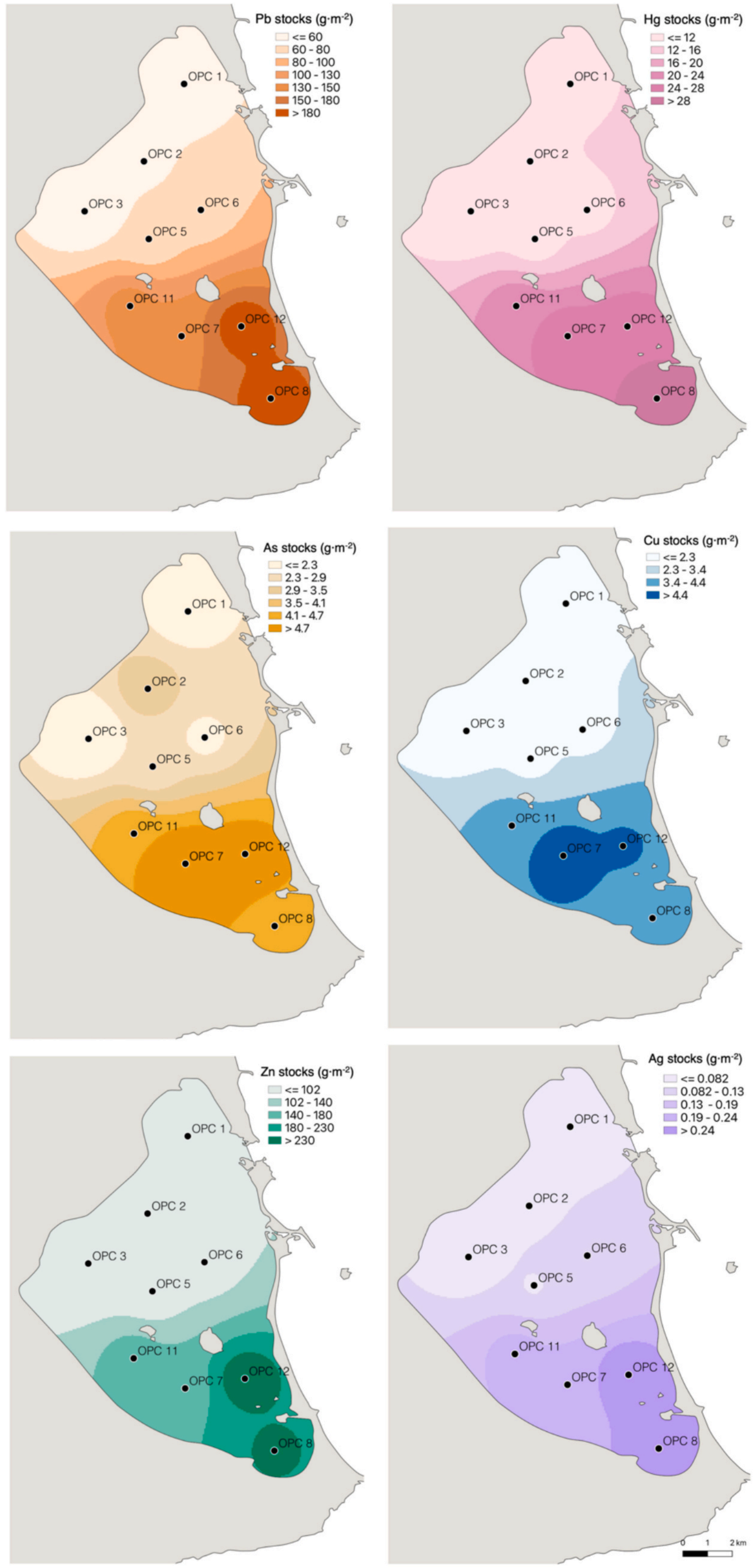

Fig. 4. Spatially interpolated sediment metal stocks of Pb, As, Hg, Cu, Zn, and Ag accumulated from 1900 ± 20 until 2020.

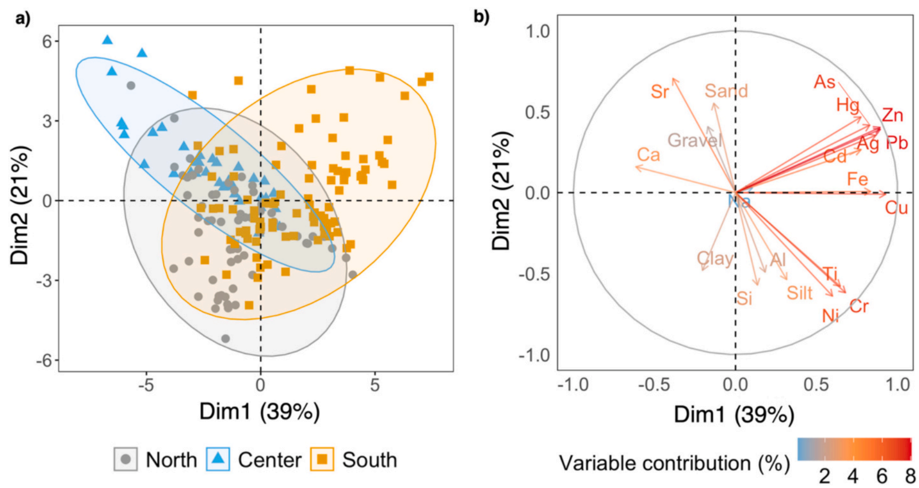

Fig. 5. Principal Component Analysis biplots for **a)** individuals (core sections) and **b)** the sediment variables. Sections included dates from 1900 to 2020 (i.e., the sections within the excess ^{210}Pb horizon). Data used includes grain size and metal concentration results. Superimposed on the **a)** plot are the confidence ellipses (95 %) for the lagoon sectors: north (grey circles), center (blue triangles), and south (orange squares). In the **b)** plot, the variable contributions (%) represent the percentage that each variable contributes to the cumulative (total) variance. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Natural and anthropogenic factors have influenced the sediment composition and changes in metal concentrations in the Mar Menor coastal lagoon over the past century. In the northwest of the lagoon, the sand-rich deeper sections of cores OPC2 and OPC3 suggested a higher influence of Las Encañizadas sandy materials in the past. The increase in silt content in recent (after 1910) core sections coincides with an increase in metal concentrations (See Section 3.1) and with the developing period of industrial mining. Therefore, there might be a confluence of factors in the north that explain this increase in sedimentation of finer materials from the late 19th century onwards.

Cores from the north and central sectors showed lower sedimentation rates and thinner layers of mining contamination (upper 15 cm at most), with lower metal concentrations and less variability, as indicated in the PCA biplots (Fig. 5). Southern cores, more affected by mining-derived materials, spanned over a larger biplot area (Fig. 5 a) and therefore showed a more diverse influence of the different variables. Additionally, these cores exhibited higher differences in metal concentrations before and after industrial mining. For instance, core OPC8 showed concentrations of 120 mg·kg^{-1} for Pb before industrial mining, which increased to 3400 mg·kg^{-1} after industrial mining began. Conversely, core OPC1 (in the north) presented concentrations of 43 mg·kg^{-1} for Pb in undated (before ~1900), deep sediments, which increased to 870 mg·kg^{-1} after the onset of industrial mining.

The PCA generated two underlying variables that explained the variability of our data. Dim1 differentiated mining-derived materials (positive values) from other sedimentary sources (negative values). Dim2 differentiated between terrestrial and marine sediment inputs as it reflected grain size variations (a larger proportion of finer materials for negative values) that could be linked to differences in terrestrial as opposed to marine inputs. Ca, Sr, sand, and gravel were probably marine-sourced (Abu-Zied et al., 2021) –either autochthonously produced or by marine inputs through the channels of the sand bar.

This analysis indicated that Pb, Zn, Ag, Cd, Fe, and As, found in ores historically mined in the Sierra Minera de Cartagena-La Unión, were associated with finer, terrigenous materials and mining runoff. Other metals not directly exploited in the mining area, such as Hg, also shared this origin. Ni, Cr, Al, Si, and Ti, were mainly influenced by the deposition of finer sediments but were not clustered with mining-derived materials, indicating that they were primary lithogenic runoff components. Cu presented an intermediate situation between the group of mining metals and the lithogenic runoff components, suggesting multiple sources.

4.2. *Metal contamination is caused by industrial mining and recent anthropogenic contributions*

The metal concentrations in the Mar Menor coastal lagoon sediments reflected a complex interplay of historical and recent contamination sources, which included mining and other anthropogenic activities. Mining metal concentrations followed similar temporal trends across all sectors of the lagoon, closely tied to the history of mining in the Sierra Minera de Cartagena-La Unión. Their concentrations presented significant increases linked to the onset of industrial mining before and at the start of the 20th century. This increase in metal concentrations around 1900 has been observed in similar coastal systems (Buckley et al., 1995). Maxima occurring in the first half of the 20th century. The significant decrease in most metal concentrations around the 1950s and 1960s, particularly in the central and southern sectors, can be explained by the 1955 ban on premeditated mining spills into the lagoon. After the ban, torrential rain runoff from the mining area still delivered metals to the lagoon (María-Cervantes et al., 2009; Marín-Guirao et al., 2005a). Similarly, to Pb and Zn, As concentrations increased due to the development of large-scale mining activities, but recent higher concentrations could be related to continuous inputs of this metal even after the ceasing of mining activities. Inputs of As could be linked to the sensitivity of this metal to redox conditions (Zitoun et al., 2024), facilitating As leaching from soils after acidification and subsequent delivery to the lagoon through stream discharge (Martínez-López et al., 2020).

While mining has been a major contributor to metal contamination, other anthropogenic activities have emerged as significant sources in recent decades. For Hg, mining was the main contributor to its enhanced concentrations in sediments throughout most of the 20th century. However, high concentrations in recent sediments could be linked to non-mining sources, especially in the northern sector. Its proximity to large population centers, such as San Pedro del Pinatar and San Javier, may contribute to Hg contamination due to broad anthropogenic sources. Furthermore, the presence of an airport and a water treatment plant in San Javier and a landfill in San Pedro —infrastructures related to Hg emissions (Fang et al., 2018) — could further increase the supply of Hg to lagoon sediments. Other potential sources for Hg include agricultural wastes (Ruiz-Fernández et al., 2009).

Urban expansion can explain the increase in Cu concentrations at the start of the 21st century. Antifouling paints are the most probable source of Cu (Costa et al., 2016; Readman, 2006), especially near the large ports of Lo Pagán and La Manga, and after the enlargement of El Estacio

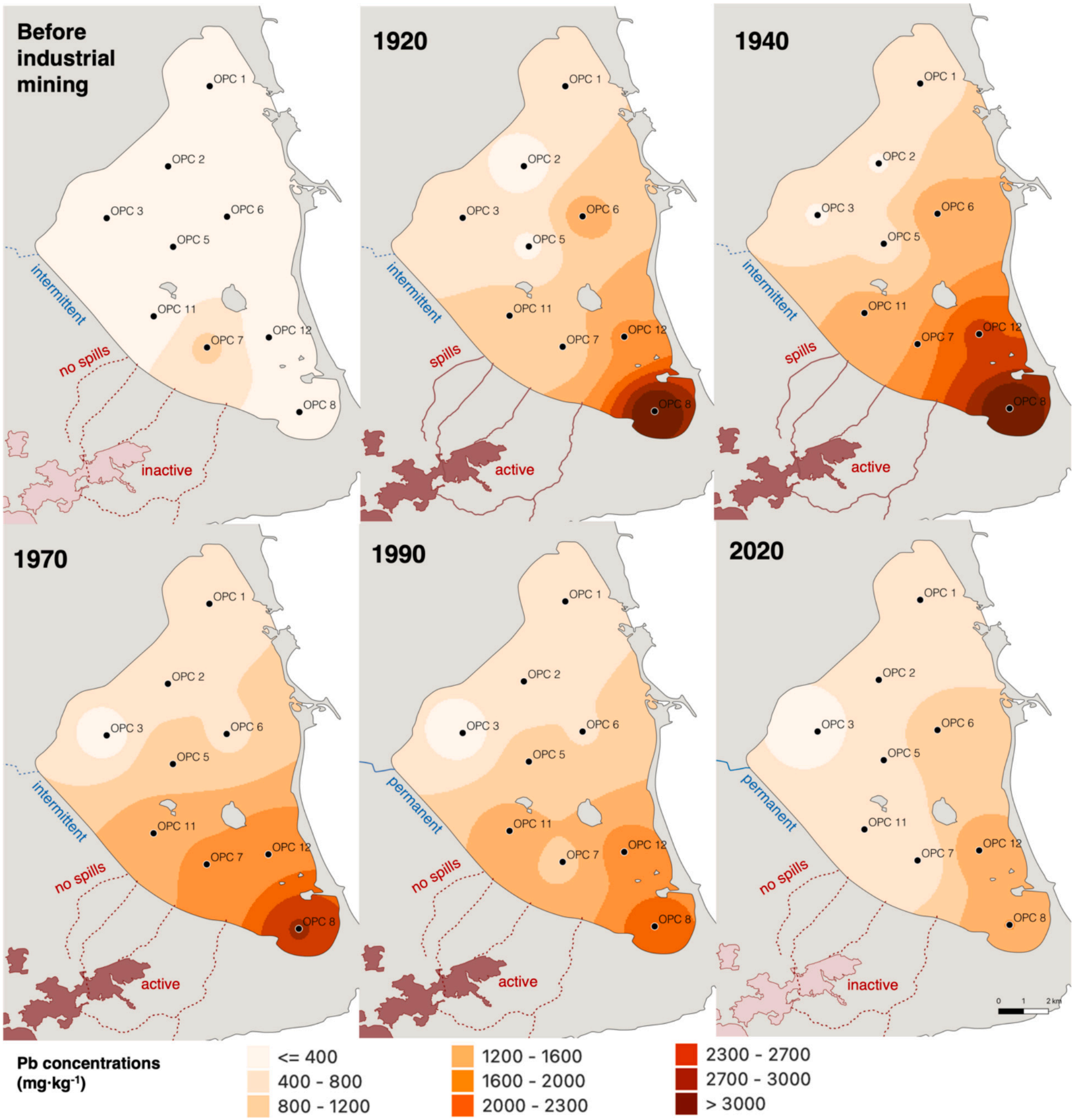

Fig. 6. Pb surface concentrations (mg·kg^{-1}) at different periods. The dotted line in the ephemeral streams affected by mining indicates a minor influx of mining debris into the lagoon, either from preindustrial mining or due to the spilling ban. Similarly, the solid line in the Albujón watercourse signifies its permanency or intermittence. The clear and dark mining sites areas indicate the lack or presence of industrial exploitation.

channel for sailing in 1973. Additional sources of metals to the central and southern sectors could include fertilizer application, which can trigger the remobilization of metals in soils due to acidification (Krahforst et al., 2022). Although Ag accumulation through time is mainly controlled by the mining industry activities, there were increases in the 1980s to the 2000s in the northern and southern basins that could be indicative of sewage releases (García-Pintado et al., 2007) due to the use of Ag nanoparticles in antibacterial consumer goods (Gallon and Flegal, 2015; Ravizza and Bothner, 1996).

As a consequence of the temporal variability of metal inputs and accumulation rates, metal concentrations in surface sediments of the lagoon have changed during the last 120 years. The characterization of the temporal evolution of surface sediments is relevant as they are a good representation of the metal concentrations that were readily available for benthic and planktonic organisms and prone to be mobilized to the water column. This is particularly relevant for those metals presenting high concentrations, such as those derived from mining activities. As shown before, metal-enriched sediments have consistently deposited in all sectors of the lagoon, but more prominently in the south, closer to the mouths of the streams from the mining area, and where there is a circulation gyre that enhances sediment accumulation.

Focusing on mining-derived metals and using Pb as an example (Fig. 6), concentrations in surface sediments of the lagoon were relatively low and showed minor spatial variability before industrial mining. At this period, the area in front of the ephemeral streams draining the mining area (Sierra Minera de Cartagena-La Unión), where OPC7 is located, already presented higher concentrations than the rest of the lagoon. These higher concentrations are probably caused because these

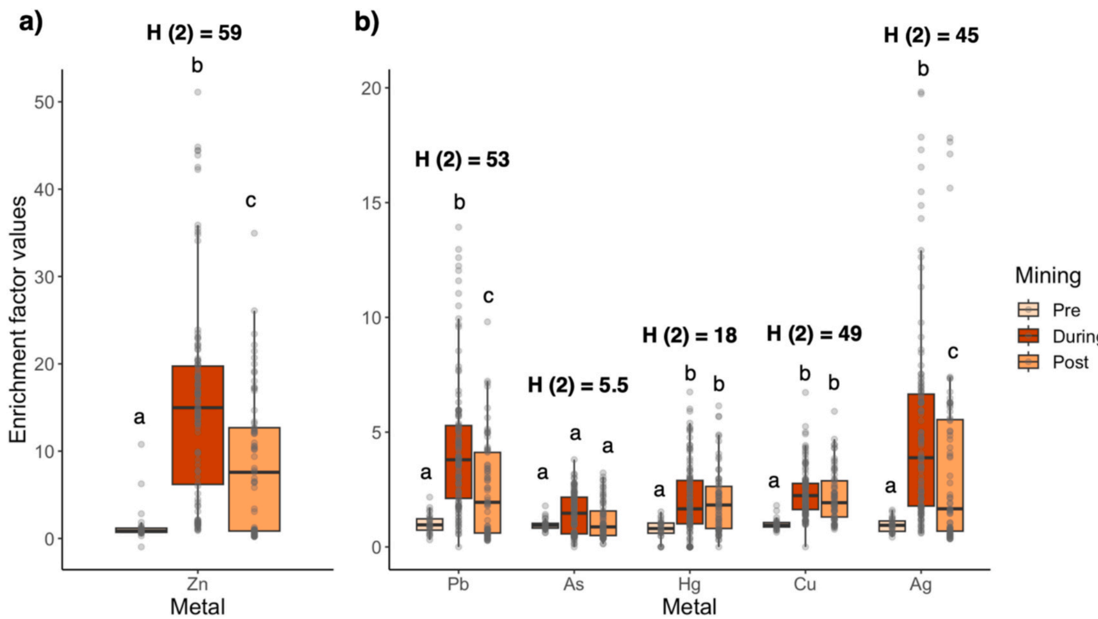

Fig. 7. Enrichment factor values for a) Zn and b) Pb, As, Hg, Cu, and Ag in the periods previous (before the second half of the 19th century), during (from then to 1990), and after mining closure (post-1990), index H from Kruskal-Wallis analysis (for Pb, Hg, Cu, Zn and Ag $p < 0.01$; for As p = 0.14) and the results of Dunn's test (adjusted p < 0.01) for the pairwise differences. The columns with different letters (a, b, c) are significantly different from one another. Box comprises the data between the 25th and the 75th percentiles. Whisker size represents 1.5·SD.

watercourses run through the Sierra, which contains volcanic materials rich in minerals that were historically exploited. The period with the highest metal concentrations, which also contributes more to the accumulation of metal stocks, occurred during the 1920s and 1940s (Fig. I1 and Table I1). After the establishment of industrial mining, the highest concentrations were always found at the southernmost edge, due to the circulation gyre and the proximity of mining-derived inputs. As observed in the distributions in the 1970s and 1990s, surface metal concentrations in the lagoon started decreasing, particularly in the south, as a consequence of the mining spilling ban in 1955. This decrease began to affect the rest of the lagoon, especially the area in front of the ephemeral streams affected by mining (OPC7 and OPC11) after the 1990s. Contamination has also preferentially spread along the eastern coast of the lagoon, as shown in the maps for 1920, 1940, and 2020 (Fig. 6). The current flowing to the northeast, which runs between the two main islands, could drag these materials, subsequently depositing them along the southward current flowing along La Manga (Fig. 1). In contrast, the north-western region, where cores OPC2 and OPC3 are located, might have been more influenced by ephemeral streams and Mediterranean inputs that do not supply mining-related metals.

4.3. Metal concentrations surpass sediment quality guidelines

Enrichment factors (EF) for Pb, As, Hg, Cu, Zn, and Ag before, during, and after mining industrial exploitation are shown in Fig. 7. Values were, on average, higher than 1.5 for the six metals during and after industrial mining, indicating accumulation of anthropogenic inputs (Gao and Li, 2012). The metal that showed the major enrichment was Zn, particularly during industrial mining times, followed by Ag. After the closure of all mining activities, average enrichment factors for Pb, Zn, and Ag decreased, with As being the closest to preindustrial values (EF close to 1). EF values for Pb, Zn, and Ag were significantly different when comparing every defined period ($p < 0.01$). For As, enrichment values did not show significant differences between periods ($p = 0.14$). This further demonstrates that factors other than the operational activity status of mining, such as recent leaching from mining watercourses, also play a role in the enrichment of this metal. Regarding Hg and Cu, the industrial mining and the post-mining periods were not significantly different, whereas the preindustrial mining values differed significantly when compared to mining and post-mining periods (both $p < 0.01$). This, again, underpins additional anthropogenic sources of Hg and Cu after mining cessation.

The measured metal concentrations were compared with sediment quality guidelines (SQG) to estimate their potential ecotoxicological effects. The chosen SQG are the Threshold Effect Level (TEL) and Probable Effects Level (PEL) (Macdonald et al., 1996), which are, respectively, the sediment metal concentration below which adverse effects rarely occur in benthic organisms and the concentration at which

Table 3
Reported metal concentration ranges (mg·kg^{-1}) for coastal lagoons and bays.

Site	Sample type	Pb	Hg	As	Cu	Zn	Ag	References
Mar Menor - this study	**Sediment cores**	**10–3400**	**0.01–0.53**	**4.6–100**	**1.1–50**	**2.6–3700**	**0.020–2.3**	**This work**
Al-Shuaiba lagoon	Grab sampler	2–270	–	–	3–200	3–400	–	Abu-Zied et al. (2021)
Altata-Ensenada lagoon system	Grab sampler	46–290	–	–	5.6–64	19–170	1.6–7.0	Green-Ruiz and Páez-Osuna (2001)
Berre Lagoon, MedSea	Grab sampler	18–82	0.15–0.40	4–10	11–48	50–150	–	Accornero et al. (2008)
Laizhou Bay	Grab sampler	16–32	–	8.1–17	7.0–34	46–91	–	Dang et al. (2021)
Nakaumi lagoon	Grab sampler	12–35	–	4–19	8–51	42–210	–	Ahmed et al. (2010)
Nador lagoon, MedSea	Sediment cores	11–300	–	4–76	4–470	4–1200	–	Bloundi et al. (2009)
Terminos lagoon	Sediment cores	6.9–19	–	0.9–11	13–23	40–66	–	Ontiveros-Cuadras et al. (2024)
Venice industrial site, MedSea	Sediment cores	41–930	1.2–48	7–130	–	100–8300	–	Bellucci et al. (2002)
Vistula Baltic coast lagoon	Grab sampler	24–80	–	4–20	18–32	76–180	0.37–0.81	Szefer et al. (1999)
Xola-Paramán lagoon	Sediment cores	9–15	–	1.8–5.6	6.1–28	37–100	–	Ontiveros-Cuadras et al. (2021)

adverse effects are frequently observed in benthic communities. These values are indicated in Fig. 2 along with the metal concentrations in the sediment profile.

Preindustrial mining concentrations of Pb, Hg, As, Cu, and Zn already exceeded TEL values, indicating pre-contemporary potentially toxic fluxes of these metals to the lagoon attributed to inputs from the watershed and in agreement with previous observations from Pérez-Ruzafa et al. (2023). Concentrations of Pb, As, and Zn during the pre-industrial period even surpassed PEL values, although As concentrations only reached this level before the 20th century in the north and center regions. During the industrial mining era, these metal concentrations further increased, reaching up to 30, 6, and 14 times PEL values for Pb, As, and Zn, and 4 and 3 times TEL values for Hg and Cu, respectively. Current concentrations are still above PEL for Pb, As, and Zn and above TEL for Hg and Cu. For Hg, surface concentrations may soon show non-toxic values if the latest decreasing tendencies persist. Even though a general, relatively low bioconcentration factor has been found in most lagoon organisms, indicating effective metal retention by sediments (Pérez-Ruzafa et al., 2023), such high concentrations could have negative impacts on biota if their bioavailability changes.

4.4. Mar Menor sediments present some of the highest metal concentrations in coastal enclosed ecosystems

Metal sediment contamination in the Mar Menor coastal lagoon is notably high compared to other reported values in lagoons within the Mediterranean region and on a global scale (Table 3). Pb maximum concentrations in this study exceeded those reported in many similar ecosystems, including the industrial area of the Venice lagoon (Bellucci et al., 2002). This location was the only one that presented higher maximum As, Zn, and Hg values than Mar Menor. Cu maximum concentration in Mar Menor was of a similar magnitude to those reported in other lagoonal ecosystems like Berre (France) —affected by industry— and Naukaumi —affected by mining— (Japan) (Ahmed et al., 2010; Ontiveros-Cuadras et al., 2024), but notably lower than Nador and Al-Shuaiba lagoons, which are impacted by a combination of mining and urban pollution (Abu-Zied et al., 2021; Bloundi et al., 2009). For Pb, As, and Zn, Naukami presented lower sediment metal concentrations than Mar Menor (Ahmed et al., 2010). Ag maximum concentration was two times higher in the Mar Menor coastal lagoon than in the Vistula Baltic coastal lagoon, impacted by industry (Szefer et al., 1999). Lagoons affected by agricultural contamination also exhibit generally lower metal concentrations in their sediments than Mar Menor (Green-Ruiz and Páez-Osuna, 2001; Ontiveros-Cuadras et al., 2021), highlighting the polluting potential of industrial mining.

4.5. Enhanced resuspension could remobilize accumulated metals to the water column

Despite generally decreasing trends in metal concentrations in recent sediments, it is crucial to consider the substantial accumulation of metals in a vulnerable ecosystem such as the Mar Menor coastal lagoon. The mobility and potential ecological effects of these metals in sediments are influenced by various factors including grain size (Horowitz, 1985), pH (Bourg and Loch, 1995), dissolved oxygen (Abu-Zied et al., 2021), redox potential (Williams, 1992), iron-manganese oxides (González et al., 2007), organic matter content, salinity (Förstner et al., 1986), and the influence of living organisms (Souza et al., 2015).

Metals can become available for biological uptake when they detach from mineral oxides or clay coatings, a process enhanced by the oxidation of anaerobic sediments (Chapman et al., 2011). This phenomenon is particularly notable at the oxic-anoxic interface, which plays a dominant role in the fluxes of trace metals from sediments to the water column (Salomons, 1987). During resuspension episodes, metals can be exposed to oxygenated water layers and become more accessible for uptake (Caetano et al., 2003; Santos-Echeandía et al., 2023). Additional mechanisms such as diffusion, porewater exchange, and submarine groundwater discharge through sediments can also contribute to mobilizing metals from lagoon sediments to the water column (Alorda-Kleinglass et al., 2019; Martínez-Soto et al., 2016).

Albeit metal concentrations in Mar Menor's surface sediments exceed sediment quality guidelines for metals like Pb, Hg, As, Cu, and Zn, a general, relatively low bioconcentration factor is found in most lagoon organisms, indicating effective metal retention by sediments (Pérez-Ruzafa et al., 2023). The rise in organic matter fluxes to Mar Menor sediments due to agricultural activities and urban sewage discharge could have impacted the lagoon sediment's ability to absorb As and contribute to its release (Martínez-López et al., 2020) while retaining other metals in the sediments. This retention is presently preventing their bioavailability and mitigates potential ecotoxicological impacts (Zhang et al., 2017). However, there is evidence that in the event of strong winds, resuspension increases metal concentration in the water column for both the dissolved and particulate fraction (Santos-Echeandía et al., 2023).

Mediterranean coastal lagoons are particularly vulnerable habitats to future global change scenarios. The expected rise in extreme precipitation events (like the ones produced by cut-off lows in the region), more frequent in recent years in the western Mediterranean due to climate change (Benabdelouahab et al., 2020; Kyselý et al., 2012), could lead to more recurrent runoff-induced episodic discharges from contamination sources. Additionally, they could increase resuspension (Fong et al., 2020), especially in shallow environments like coastal lagoons (Tang et al., 2020). In Mar Menor's case, this could affect the most polluted sediments from the last decades (Palanques et al., 2020) and increase the availability of sediment-bound metals upon exposure to oxygen, as shown in Bancon-Montigny et al. (2019), potentially affecting the biota. Additionally, expected sea level rise from climate change could contribute to flooding the contaminated watershed soils and increase metal mobilization to the water column (Izaditame et al., 2022).

The 2016 and 2019 extreme precipitation events in the Mar Menor, could have contributed to delivering mining-polluted sediments by runoff and conveyed high organic matter and nutrient loads to the lagoon, which could contribute to metal retention but at the same time trigger new eutrophication peaks. Additional challenges, such as mass mortality episodes in benthic macrophyte meadows, could hamper their contribution to sediment-retaining processes and further increase resuspension events. Such delicate equilibrium in an already remarkably vulnerable ecosystem requires more detailed studies, especially focusing on resuspension risk and properly planned metal contamination monitoring programs.

5. Conclusions

The development of the mining industry in the Sierra Minera de Cartagena-La Unión during the 19th and 20th centuries led to the release of large quantities of metals that reached Mar Menor, Europe's largest saltwater lagoon. As a consequence, its sediments are enriched in Pb, As, Zn, Hg, Cu, and Ag in concentrations that surpass the toxicity thresholds and are generally higher than values reported in other marine coastal enclosed ecosystems affected by metal contamination. The southern sector of the lagoon, closest to the mining mountain range, has accumulated the highest metal stocks. The mining industry is responsible for the increase in metal concentrations everywhere in the lagoon from the start of the 20th century until the 1950s. Since then, the general decline in metal concentrations can be largely attributed to the 1955 ban on mining spills and the cessation of mining activities in 1991. After mining activities ceased, leaching from the watershed soils continued to supply metals to the sediments. Additional anthropogenic sources such as antifouling paints and sewage discharges also delivered Hg, Cu, and Ag to the lagoon. Nowadays, surface metal concentrations still exceed sediment quality guidelines, and although sediments seem to be effectively retaining these metals, changes in the lagoon's conditions could alter this situation and promote their mobility. Climate change and eutrophication episodes might increase the resuspension risk of historically accumulated contaminated sediments, favoring their bioavailability, and endangering biota. Many marine ecosystems affected by historical metal contamination and vulnerable to resuspension risks could endure new effects of this pollution. Therefore, studies focused on metal contamination from the source and remobilization perspectives are crucial for developing future management plans for marine coastal ecosystems.

CRediT authorship contribution statement

Irene Alorda-Montiel: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Valentí Rodellas:** Writing – review & editing, Supervision, Resources, Project administration, Investigation, Funding acquisition, Conceptualization. **Ariane Arias-Ortiz:** Writing – review & editing, Supervision, Resources, Methodology, Conceptualization. **Albert Palanques:** Writing – review & editing, Conceptualization. **Andrea G. Bravo:** Writing – review & editing, Conceptualization. **Júlia Rodriguez-Puig:** Writing – review & editing, Investigation. **Aaron Alorda-Kleinglass:** Writing – review & editing, Investigation. **Carlos Green-Ruiz:** Writing – review & editing, Conceptualization. **Marc Diego-Feliu:** Writing – review & editing, Investigation. **Pere Masqué:** Writing – review & editing, Conceptualization. **Javier Gilabert:** Writing – review & editing, Project administration, Funding acquisition. **Jordi Garcia-Orellana:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2025.118347.

Data availability

Data is available at:

Replication Data for: A Century of Sediment Metal Contamination of Mar Menor, Europe's Largest Saltwater Lagoon (Original data) (CORA)

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