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Natural and anthropogenic decoupling of deep-sea mercury records from global emission trends

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Abstract

Mercury (Hg) concentrations preserved in marine sediments are widely used to reconstruct the history of anthropogenic contamination. However, whether deep-sea sediment records reflect recent changes in global Hg emissions remains unclear. Using 26 dated sediment cores from submarine canyons and adjacent anthropogenically undisturbed environments across the northwestern Mediterranean, we reconstructed more than a century of Hg accumulation. Hg concentrations in undisturbed environments continued to increase despite recent changes in global Hg emissions. By contrast, concentrations declined after the 1970s only along submarine canyon axes affected by bottom trawling. These declines resulted from dilution caused by trawling-induced resuspension and redeposition of older sediments. Our findings show that deep-sea Hg records are shaped by two contrasting processes: delayed oceanic Hg cycling, which prolongs the legacy of historical contamination, and bottom trawling, which overprints the natural sedimentary signal through sediment remobilization. Consequently, deep sediment Hg concentrations can be decoupled from recent atmospheric Hg emissions.

Introduction

Deep-sea sediments provide one of the most important natural archives for reconstructing past environmental change. Because they accumulate particles delivered from the atmosphere, continents, and the overlying water column, they preserve long-term records of natural variability and anthropogenic disturbance. Consequently, sedimentary records have been widely used to reconstruct historical trends in contaminant inputs, evaluate the effectiveness of environmental regulations, and assess the long-term evolution of the global biogeochemical cycles¹.

Mercury (Hg) is among the most important contaminants archived in marine sediments because of its persistence, toxicity, and global atmospheric transport. Since the Industrial Revolution, anthropogenic activities have greatly increased Hg emissions to the atmosphere, resulting in enhanced Hg deposition to the ocean and increasing Hg accumulation in marine sediments²⁻⁹. However, the evolution of anthropogenic Hg emissions during the past decades has been complex^{1,10-12}. Although emissions associated with commercial Hg use declined after the 1970s and atmospheric emissions decreased in Europe and North America^{11,13} global Hg emissions have followed a different trajectory owing to increasing emissions from other regions¹² together with the continued recycling of legacy Hg stored in environmental reservoirs^{10,14,15}. Atmospheric global emissions increased until about 1980, remained approximately stable over the following two decades, and increased again after 2000¹². Whether these recent changes are faithfully recorded in deep-sea sedimentary archives remains unclear.

Interpreting deep-sea Hg records is further complicated by the fact that the transfer of Hg from the atmosphere to the seafloor is not instantaneous. Following atmospheric deposition, Hg is extensively recycled through physical, chemical, and biological processes within the ocean before its eventual burial in deep-sea sediments^{8,16}. In addition, anthropogenic activities can modify sediment transport and deposition after particles have reached the seafloor.

Among these activities, bottom trawling has become one of the most widespread human disturbances affecting continental margins¹⁷. Its impact intensified from the 1970s onward, when increasingly powerful fishing vessels equipped with heavy otter doors began operating at water depths of up to 900 m¹⁸⁻²⁰. Although bottom trawling is known to alter sediment accumulation and geochemical records¹⁹⁻²³, its influence and implications on century-scale reconstructions of Hg contamination have not been evaluated.

Here, we reconstruct more than a century of Hg accumulation using 26 dated sediment cores collected from submarine canyons, canyon flanks, the open continental slope, and intra-slope basin environments across the northwestern Mediterranean (Fig. 1). By comparing environments affected and unaffected by bottom trawling, we distinguish the influence of sediment remobilization from Hg inputs. We show that deep-sea sediments from anthropogenically undisturbed environments continue recording the long-term legacy of anthropogenic Hg contamination despite recent changes in global Hg emissions, whereas bottom trawling produces an artificial decline in Hg concentrations through sediment remobilization and dilution. Our results demonstrate that delayed oceanic Hg cycling and anthropogenic alteration of sediment dynamics must both be

considered when interpreting deep-sea sedimentary records of historical contamination.

Results

Study area and sedimentary settings

The 26 dated sediment cores were collected in the Catalan and Valencia continental margins. The northern and central Catalan margin is incised by submarine canyons that intercept sediment exported from the narrow continental shelf (Fig. 1B), whereas the southern Catalan and Valencia margins are characterized by a wider shelf where canyons are disconnected from the coastline (Figs. 1A, C). Since the 1970s, bottom trawling has enhanced sediment resuspension and downslope transport along the axes of the canyons from the northern and central Catalan margin^{17-22,24}, whereas most of canyon flanks, the open slope, intra-slope basins, and the southern margin provide comparatively sedimentary environments undisturbed by anthropogenic activities.

Chronology and sediment accumulation

Before the 1970s, sediment accumulation rates were highest in the Palamós, Blanes, Besòs and Foix canyons, whereas the remaining canyons and undisturbed sites exhibited substantially lower values (Table 1). After the 1970s, SARs increased by two to tenfold in the Palamós, Blanes, and Foix canyons and also in the shallower cores from the Arenys, Besòs, and Morràs canyons

leading to SARs of 0.3 to 4 cm yr⁻¹. These increases in sediment accumulation results from the chronic resuspension and downslope transport of sediments remobilized by bottom trawling^{17, 20-22,18,19,25}.

In contrast, sediment cores from sites unaffected by bottom trawling, including the Columbretes submarine canyons, the Foix and Palamós canyon flanks, the Blanes open slope, and the Valencia basin, showed no significant post-1970 increases in the sedimentation rates, maintaining consistently low values of 0.06-0.18 cm·yr⁻¹ throughout the sediment record, with the exception of the Palamós canyon flank, which had a higher sedimentation rate of 0.5 cm·yr⁻¹ (Table 1).

Historical evolution of Hg concentrations

Hg concentrations in the sediment cores increased throughout most of the twentieth century at all study sites. However, the post-1970 evolution of the Hg concentrations differed markedly between sites in submarine canyons affected by bottom trawling and adjacent undisturbed deep-sea environments. In trawled canyons of the Catalan margin, Hg concentrations generally peaked during the 1970s and subsequently declined. In contrast, Hg concentrations continued to increase towards the core top in areas unaffected by trawling-induced sediment resuspension (Fig. 2, Supplementary Fig. 1, Table 2).

Maximum Hg concentrations were remarkably similar among the canyons of the northern Catalan margin despite differences in water depth (Fig. 3; Table 2). In the central Catalan margin, the Foix Canyon displayed the highest Hg concentrations of all the studied cores, with maxima occurring during the 1970s

(Table 2). In this canyon, Hg-enriched sediments extended to water depths exceeding 1000 m, although Hg concentrations decreased progressively downslope (Fig. 3, Supplementary Fig. 1). Surface sediments at 865 m water depth still had up to 180 ng g⁻¹ of Hg in 2010. The Morràs Canyon also exhibited exceptionally high Hg concentrations, among the highest recorded in the study area (Table 2).

Pre-industrial Hg concentrations, estimated from the deepest sediments deposited before 1900, ranged from 19 to 56 ng g⁻¹, except in the Foix Canyon, where they were about 100 ng g⁻¹ (Fig. 2). Relative to these baseline values (we took 25 ng g⁻¹ in Fig. 2), peak Hg concentrations were up to an order of magnitude higher in the Foix and Morràs canyons and by approximately two- to seven-fold at the remaining sites. Present-day surface sediments still contain Hg concentrations up to three times higher than these baseline levels, indicating substantial anthropogenic enrichment throughout the study area.

Hg accumulation rates and inventories

Hg-AR increased during the twentieth century at all study sites, broadly following the historical rise in Hg concentrations (Fig. 4, Supplementary Fig. 2). Before 1900, Hg-AR generally ranged from 11 to 93 µg m⁻² yr⁻¹, although higher values (up to 441 µg m⁻² yr⁻¹) were recorded in the Foix Canyon (865 and 1365 m), the Blanes Canyon (1209 m), and the Palamós canyon flank, reflecting their higher sediment accumulation rates.

From 1900 to the 1970s, Hg-AR increased by approximately 1.5- to 4-fold across the study area. Thereafter, following the increase of bottom trawling

fishing effort and disturbance that occurred during the 1970s, contrasting trends emerged between trawled submarine canyons and adjacent undisturbed environments. Peak Hg-AR occurred during the 1970s within the trawled canyons, reaching $3500 \mu\text{g m}^{-2} \text{yr}^{-1}$ in the Foix Canyon and $2300 \mu\text{g m}^{-2} \text{yr}^{-1}$ in the Blanes and Palamós canyons, whereas Hg-AR remained below $430 \mu\text{g m}^{-2} \text{yr}^{-1}$ at sites unaffected by bottom trawling (Fig. 4, Supplementary Fig. 2; Table 2).

After the 1970s, Hg-AR decreased by 10–50% in trawled submarine canyons but continued to increase by 10–40% at undisturbed sites (Table 2; Supplementary Fig. 2). Despite declining Hg concentrations, Hg-AR remained high within trawled canyons

Although this period was substantially shorter, Hg inventories for 1970-2010, were higher than those for 1900-1970 in submarine canyons affected by bottom trawling (Fig. 5), whereas inventories changed only slightly at undisturbed sites (Table 2). The largest increases occurred in the Palamós, Blanes, and Foix canyons, particularly at water depths shallower than 1,400 m. More moderate increases were recorded in the shallowest cores from the Arenys, Besòs, and Morràs canyons, while inventories remained nearly unchanged in their deeper cores and at sites unaffected by bottom trawling. (Fig. 5; Table 2).

Discussion

Deep-sea sedimentary Hg records reflect not only Hg inputs to the seafloor but also the sedimentary processes that redistribute and bury contaminated particles after deposition. Distinguishing between these two controls is therefore

essential for interpreting marine sedimentary archives. Across most of the study area, the occurrence of remarkably similar maximum Hg concentrations despite large differences in water depth (Fig. 3) indicates that diffuse atmospheric deposition, the dominant external source of Hg to the open Mediterranean Sea^{26,27}, is the primary control on the regional Hg signal preserved in deep-sea sediments.

The Foix and Morràs canyons constitute notable exceptions to this regional pattern. In these canyons, Hg concentrations were substantially higher than elsewhere and decreased progressively with water depth (Fig. 3), indicating the local offshore transport of highly contaminated sediments from the densely urbanized and industrialized Barcelona metropolitan area through the Foix submarine canyon.^{28,29} Similar contamination gradients have been reported from other submarine canyons influenced by major metropolitan areas, such as the Oreto Canyon (Palermo) and the Cascais Canyon (Lisbon)^{23,30}, emphasizing that submarine canyons efficiently transfer locally contaminated sediments from the continental shelf to the deep sea.

Apart from this local anomaly, the divergence observed since the 1970s between environments affected and unaffected by bottom trawling is consistent with observations from other deep-sea areas. The absence of post-1970 declines in Hg concentrations can be observed in sediment cores from other undisturbed Mediterranean and northeastern Atlantic environments, including the open continental slopes adjacent to the Foix Canyon and the Ebro margin, the northern Balearic slope³¹, the Gulf of Lions continental rise and abyssal plain¹, the Gulf of Palermo³² and the Extremadura Spur³⁰. In contrast, declining Hg concentrations have been observed in cores from other Mediterranean

submarine canyons affected by bottom trawling, such as the Cap de Creus³¹, Oreto, Eleuterio and Arenella submarine canyons²³.

Chronic bottom trawling along submarine canyon flanks resuspends large volumes of recently deposited shelf sediments that are transported downslope as dilute sediment gravity flows^{24,33}. During transport, hydraulic sorting preferentially deposits coarser particles while finer fractions remain in suspension and are carried farther downslope³⁴⁻³⁶. This process progressively increases silt content (Supplementary Fig. 3, 4), and enhances sediment accumulation rates along canyon axes by up to an order of magnitude (Table 1)^{19, 20}. At the same time, repeated erosion and redeposition mix Hg-enriched surface sediments with older, less contaminated material, lowering the Hg concentration of the redeposited sediment.

However, although sediment Hg concentrations declined after the 1970s (Fig. 2), Hg accumulation rates and inventories remained substantially higher than before 1970 in the Palamós, Blanes, and Foix canyons (Fig. 5, Supplementary Fig. 2) because the marked increase in sediment accumulation more than compensated for the decrease in sediment Hg concentrations.

The persistence of increasing Hg concentrations in undisturbed environments has broader implications for interpreting marine contaminant archives. None of the major changes in anthropogenic Hg emissions since the 1970s are yet reflected in the deep-sea sediment records examined here. Instead, these sediments continue to record the cumulative increase in anthropogenic Hg throughout the twentieth century, indicating that the response of deep-sea sediments to changing Hg emissions is substantially delayed³⁷.

This delayed response is consistent with the present understanding of the global Hg cycle. Anthropogenic Hg is transferred gradually from surface waters to the deep ocean through particle scavenging, biological recycling, and large-scale circulation^{10,38}. The large oceanic Hg reservoir buffers and delays the propagation of changing atmospheric Hg inputs because previously accumulated Hg continues to circulate and be scavenged onto sinking particles. Continued remobilization of legacy Hg from soils, river catchments, shelf sediments, and other historical reservoirs further smooths temporal variations in atmospheric inputs^{10,14,15,39}. Collectively, these processes explain why deep-sea sediments preserve the long-term legacy of anthropogenic Hg contamination rather than responding immediately to recent changes in atmospheric emissions.

Our observations suggest that deep-sea sedimentary Hg records primarily reflect the evolving Hg inventory of the ocean, which integrates anthropogenic inputs over multi-decadal timescales before they become incorporated into deep-sea sediments. Consequently, the absence of recent changes in sediment Hg concentrations should not be interpreted as evidence that atmospheric Hg emissions have remained unchanged. Rather, it reflects the delayed response of the oceanic Hg cycle.

Bottom trawling introduces a fundamentally different control on this sedimentary archive. Whereas the slow oceanic Hg cycle delays the transmission of changes in Hg emissions to the deep sediments, bottom trawling modifies the sedimentary record itself through erosion, resuspension, mixing, downslope transport and redeposition. These processes decouple Hg concentrations and burial rates from the history of anthropogenic Hg emissions.

Taken together, our results reveal a dual control on modern deep-sea Hg records. In undisturbed environments, the slow oceanic Hg cycle produces a natural temporal decoupling from Hg emission trends, causing deep-sea sediments to integrate the long-term legacy of anthropogenic contamination. In contrast, bottom trawling introduces an anthropogenic overprint by modifying sediment transport and accumulation after deposition. Thus, the Hg signal preserved in deep-sea sediments reflects not only the history of Hg inputs but also the processes governing its transfer, redistribution, and burial at the seafloor. The conceptual model presented in Fig. 6 summarizes these contrasting pathways and illustrate how natural oceanic processes and human-induced sediment disturbance interact to shape the Hg record preserved in deep-sea sediments.

Although demonstrated here for Hg, these findings are likely to extend to other particle-reactive contaminants and geochemical proxies preserved in marine sediments. More broadly, they show that interpreting deep-sea sedimentary archives requires distinguishing between processes that control contaminant delivery to the seafloor and those that subsequently can modify the sedimentary record.

Methods

Study area

The study region comprises the Catalan margin in the north of the study area and the Valencia margin in the south⁴⁰. Sediments in the northern and central Catalan margin predominantly originate from small rivers, including the Muga,

Fluvià, Ter, Tordera, Besós, Llobregat, Foix and Francolí, Rivers, while the southern Catalan margin and the Valencia margin receive sediment from the larger Ebro River (Fig. 1). River-derived sediments and associated contaminants accumulate in shelf mud belts and are transported southward along the margin by the Northern Current and episodically exported offshore during storms and dense shelf water cascading events^{28,29,41-43}

The northern and central Catalan margins are characterized by 6 to 30 km wide continental shelves and are incised by large submarine canyons. The main submarine canyons of this margin include Palamós (also known as La Fonera), Blanes, Arenys, Besòs, Morràs, and Foix canyons (Fig. 1B). While the Palamós and Blanes canyons incise directly into the continental shelf, the other canyons reach the shelf edge, although their heads lie within 20 km of the coast. Canyon shelf incision and proximity to the shoreline are important factors for intercepting and trapping sedimentary matter and associated contaminants delivered from the continent and exported offshelf⁴⁵.

By contrast, the southern Catalan and the Valencia margins have a wider continental shelf up to 70 km with numerous submarine canyons and gullies that incise only the continental slope, far from the coastline (Fig. 1A, C), which further limits the transfer of contaminated sediments from the shelf to the slope.

The historical evolution of trace metal contamination has been studied in sediments from the Besòs and the Llobregat prodeltas^{28,45}. In addition, trace metals in sediments along the inner Catalan continental shelf have been monitored from 2000 to 2011²⁹. Conversely, the record of trace metals contamination in the deep sediments have been only studied in the Foix

Canyon²⁸ and these studies include metals such as Cr, Zn, Cu and Pb but the contamination of Hg was not assessed.

Field work

A total of 26 sediment cores were collected from the Catalan and Valencia margins at depths ranging from 500 to 2000 m during multiple oceanographic cruises mainly from 2010 to 2021 (Fig. 1, Supplementary Table 1). Twenty sediment cores were retrieved from the axes of submarine canyons, 2 from canyon flanks, 3 from the open slope and 1 from an intra-slope basin. Sampling was performed using a KC Denmark A/X multicorer, complemented at specific sites with a gravity corer to obtain longer sedimentary records. The cores were subsampled at 1 cm intervals and each section was kept frozen in sealed plastic bags until subsequent analysis.

Laboratory work

Grain size fractions were determined using a Horiba Partica LA950V2 and a Coulter LS230 particle size analyzers. Prior to analysis, sediment aliquots of 1–4 g dry weight were oxidized using 20% H₂O₂. and disaggregated using 2.5% P₂O₇⁴⁻.

For Hg analysis, the sediment samples were ground and homogenized in an agate mortar. The Hg content of the sediment samples was determined using a LECO AMA254 Mercury Analyzer following the U.S. EPA Method 7473⁴⁶. The instrument's detection limit for Hg is 0.01 ng. For every 10 samples, a blank, a

PACS-2 reference material (National Research Council of Canada) and a random-replicated sample were used for analytical quality control. The overall analytical uncertainty was below 10%.

Age models for the last century for all sediment cores were obtained from the excess ^{210}Pb concentration profiles. The concentrations of ^{210}Pb were determined through the analysis of the activity of its granddaughter ^{210}Po by alpha-spectrometry⁴⁷, assuming secular equilibrium of both radionuclides at the time of analysis. Samples were spiked with a known amount of ^{209}Po , followed by an acid digestion using an analytical microwave, and polonium isotopes were subsequently plated on silver discs. Alpha emissions were quantified using Passivated Implanted Planar Silicon (PIPS) detectors (CANBERRA, Mod. PD-450.18 A.M). Supported ^{210}Pb concentrations were determined by averaging total ^{210}Pb concentrations at the base of each profile, verified with the ^{226}Ra concentrations obtained by gamma-spectroscopy in three to five randomly selected sediment samples throughout each core. Excess ^{210}Pb concentrations were then calculated from average supported ^{210}Pb concentrations from each core.

Age models and mass accumulated rates (MAR, in $\text{g}\cdot\text{cm}^{-2}\cdot\text{yr}^{-1}$) were calculated based on accumulated dry mass to correct for sediment compaction, applying the constant flux: constant sedimentation (CF:CS) model in regions of each core that presented a monotonic decrease in excess ^{210}Pb concentrations^{48,49}. Given that biological and physical reworking of superficial sediments can alter the excess ^{210}Pb concentration profiles, which can be confounded as being produced by greater sedimentation rates, the congruency of the ^{210}Pb derived dates were corroborated for selected cores with ^{137}Cs as a time-marker⁴⁹.

Sediment accumulation rates (SAR in $\text{cm}\cdot\text{yr}^{-1}$) were calculated from MAR using the average dry bulk density of the sediment.

The average sedimentation rates were used to extrapolate the age models to 1900 where necessary, beyond the horizon of excess ^{210}Pb concentrations, assuming sedimentation rates remained constant prior to the 1900s. Mercury accumulation rates (Hg-AR) were calculated by multiplying mercury concentrations by the corresponding MAR for each dated sediment interval. Hg inventories in the sediment record accumulated from 1900 to 1970 and from 1970 to 2010 were calculated by integrating the Hg-AR over the selected time intervals.

The age models of cores from the Arenys, Besòs and Morràs canyons were studied in²⁰, while those for a core from the Foix Canyon and a core from the Palamós Canyon were published in^{19,21,50} (Table 1).

DataAvailability

The data that support the findings of this study are available from the corresponding author upon request.

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Author contributions

A. Palanques and P. Puig conceived the initial framework and were involved in data collection, data analysis and conceptualization. S. Paradis and A Calafat

were involved in data collection and data analysis. Pere Masqué participated in data analysis. Ruth Durán was involved in data collection and Visualization, A.G Bravo participated in conceptualization. All the authors were involved in writing and editing.

Competing interests

The authors declare no competing interests

Sediment Core	Core Depth (cm)	Years	MAR (g·cm ⁻² ·yr ⁻¹)	SAR (cm·yr ⁻¹)	Reference
Palamós C. 744	0-57	1970s-2011	1.17 ± 0.080	1.66 ± 0.11	This paper
Palamós C.1344	0-23	2007-2011	6.32 ± 2.710	8.55 ± 3.660	<u>This paper</u>
	23-153	1975-2007	2.52 ± 0.150	4.14 ± 0.250	
	154-269	1845-1975	0.48 ± 0.050	0.67 ± 0.070	
Palamós C.1825	0-16	2001-2011	2.00± 0.030	2.40	19
	17-37	1972-2000	0.42 ± 0.040	0.71± 0.060	
	38-47	<1972	0.20 ± 0.020	0.25 ± 0.020	
Palamós C. 1984	0-16	1976-2011	0.3 ± 0.030	0.60 ± 0.048	This paper
	16-25	1817-1976	0.05 ± 0.004	0.06 ± 0.005	
Blanes C. 637	0-55	1973-2011	1.12 ± 0.040	1.27 ± 0.040	This paper
	56-131	1616-1973	0.19 ± 0.010	0.21 ± 0.013	
Blanes C. 1063	0-92	1978-2011	1.45 ± 0.040	2.21 ± 0.070	This paper
	0-148	1902-1978	0.42 ± 0.040	0.58 ± 0.060	
Blanes C. 1209	0-41	1968-2000	1.26 ± 0.140	1.39 ± 0.150	This paper
	41-111	1871-1968	0.67 ± 0.120	0.74 ± 0.130	
Blanes C. 1405	0-43	1974-2021	0.74 ± 0.030	0.99 ± 0.040	This paper
	43-66	1901-974	0.21 ± 0.020	0.26 ± 0.020	
Arenys C. 1074	2-14	1972-2012	0.18 ± 0.010	0.30 ± 0.015	20
	14-26	1824-1972	0.06 ± 0.004	0.08 ± 0.005	
Arenys C. 1410	0-16	1834-2012	0.07 ± 0.004	0.09 ± 0.006	20
Arenys C. 1632	0-19	1803-2012	0.06 ± 0.002	0.09 ± 0.003	20
Besòs C. 810	4-25	1990-2014	0.63 ± 0.060	0.98 ± 0.080	20
	28-41	1972-1990	0.61 ± 0.040	0.92 ± 0.060	
	40-66	1903-1972	0.28 ± 0.030	0.39 ± 0.040	
Besòs C.1238	0-12	1968-2012	0.11 ± 0.002	0.20 ± 0.005	20
	15-36	1826-1968	0.12 ± 0.006	0.18 ± 0.009	
Besòs C.1487	4-18	1834-2012	0.06 ± 0.002	0.08 ± 0.003	20
Morràs C.796	0-11	1973-2013	0.18 ± 0.009	0.32 ± 0.016	20
	10-22	1870-1973	0.09 ± 0.005	0.13 ± 0.007	
Morràs C.987	3-31	1863-2013	0.14 ± 0.005	0.20 ± 0.007	20
Foix C.865	0-20	2000-2013	1.18 ± 0.080	1.75 ± 0.110	21
	20-46	1970- 2000	0.68 ± 0.020	0.96 ± 0.030	
	51-79	1899-1970	0.44 ± 0.010	0.46 ± 0.010	
Foix C.1373	0-12	1950-1994	0.26 ± 0.010	0.28 ± 0.011	51
	12-30	1816-1950	0.15 ± 0.030	0.15 ± 0.003	
Columbretes C. 504	0-29	1842-2010	0.10 ± 0.002	0.17 ± 0.033	This paper
Columbretes C. 523	0-24	1842-2010	0.08 ± 0.001	0.13 ± 0.002	This paper
València B. 1230	0-10	1841-2010	0.03 ± 0.001	0.06 ± 0.020	This paper
Foix C. F. 1361	0-16	1843-2010	0.08 ± 0.009	0.09 ± 0.010	This paper
Palamós C. F. 489	0-56	1901-2010	0.39 ± 0.020	0.50 ± 0.020	36
Blanes O. S. 952.	0-26	1868-2010	0.129 ± 0.003	0.178 ± 0.004	22

Blanes O. S. 1294	0-17	1833-2010	0.078 ± 0.002	0.094 ± 0.003	22
Blanes O. S. 1512	0-10	1834-2010	0.064 ± 0.011	0.076 ± 0.013	22

Table 1 | Mass Accumulation Rate (MAR) and Sediment Accumulation Rate (SAR) of sediment cores. Sediment cores were collected from submarine canyons, the Foix Canyon and Palamós Canyon flanks, the Blanes Canyon open slope and the Valencia Basin. Abbreviations: Canyon (C.). Canyon flank (C. F.). Open slope (O. S.). Basin (B.). Numbers in the sediment cores are water depth

Sediment Core	Hg ng g ⁻¹				Hg-AR (µg m ⁻² y ⁻¹)				Hg Inv. (µg cm ⁻²)			Silt (%)		
	1900	1970s	2010	<i>Δ 2010-1970</i>	1900	1970s	2010	<i>Δ 2010-1970</i>	1900-1970	1970-2010	<i>Δ >1970-<1970</i>	Silt 1970s	Silt 2010	<i>Δ 2010 -1070</i>
Palamós C. 744		112.8	109.8	-3.9		1320.3	1285.1	-35.2	<1.2	4.4	<3.2	71.6	80.2	8.6
Palamós C.1344	34.0	90.3	57.5	-32.8	163.2	2275.6	1450.0	-825.6	2.0	5.8	3.7	68.2	80.8	12.6
Palamós C.1825	40.0	125.0	78.2	-46.8	72.0	1761.4	1564.2	-197.2	1.1	2.5	1.5	68.5	73.6	5.1
Palamós C. 1984	46.6	120.5	94.0	-26.5	23.3	458.0	357.2	-100.7	0.3	1.2	1.0	70.7	79.0	8.3
Blanes C. 637	40.9	144.6	85.7	-58.9	79.8	1619.2	959.3	-659.9	1.1	4.3	3.2	60.8	84.1	23.3
Blanes C. 1063	29.5	91.7	74.5	-17.2	123.8	1329.9	1080.5	-249.4	2.5	5.3	2.8	68.3	79.4	11.1
Blanes C. 1209	69.0	108.0	74.0	-34.0	462.6	1354.9	858.4	-496.5	2.6	5.2	2.6	69.4	79.6	10.2
Blanes C. 1405	41.3	97.2	79.0	-18.2	103.1	720.0	585.3	-134.6	0.7	2.1	1.4	67.2	76.0	8.8
Arenys C. 1074	34.5	113.6	107.3	-6.4	21.0	200.0	188.8	-11.2	0.3	0.9	0.6	71.1	80.3	9.2
Arenys C. 1410	53.9	124.5	113.0	-11.5	35.6	82.2	74.8	-7.4	0.3	0.4	0.1	72.3	82.0	9.7
Arenys C. 1632	70.0	125.6	109.5	-16.1	36.5	76.6	66.8	-9.8	0.3	0.4	0.1	74.8	85.8	11.0
Besòs C. 810	72.0	143.8	92.8	-51.1	201.6	877.5	584.5	-293.0	1.8	3.1	1.3	69.9	79.1	9.2
Besòs C.1238	92.1	132.6	102.6	-30.0	108.6	145.9	112.9	-33.0	0.8	0.6	-0.2	74.2	87.7	13.5
Besòs C.1487	61.5	151.8	126.3	-25.6	35.0	86.5	72.0	-14.6	0.6	0.5	-0.1	66.8	76.4	9.6
Morràs C.796	71.3	225.9	106.5	-119.4	64.2	415.7	196.0	-219.6	0.7	1.0	0.3	73.5	83.0	9.5
Morràs C.987	55.6	136.2	112.3	-23.9	71.7	175.7	144.9	-30.8	0.8	0.8	0.0	71.3	82.6	11.3
Foix C.865	166.0	497.3	184.2	-313.1	730.6	3481.2	2173.3	-1307.9	7.0	9.8	2.7	77.0	86.6	9.6
Foix C. 1373	142.6	293.0			213.9	739.2			1.6	>3.9	<2.3	69.0		
Columbretes C. 504	35.6	87.6	109.0	21.4	36.4	89.8	111.8	22.0	0.4	0.5	0.1	68.8	70.6	1.8
Columbretes C. 523	61.6	127.4	150.0	22.6	49.6	102.5	120.8	18.2	0.5	0.5	0.1	72.1	74.5	2.4
Valencia B. 1230	61.0	91.6	123.8	32.2	20.7	31.2	42.1	10.9	0.2	0.2	0.0	76.3	77.0	0.7
Foix C. F. 1361	48.0	99.0	110.0	11.0	39.0	82.0	91.0	9.0	0.3	0.6	0.3	75.3	76.0	0.7
Palamós C. F. 489	61.0	109.7	127.5	17.8	237.9	427.9	497.4	69.5	3.1	3.0	0.0	67.8	68.6	0.8
Blanes O. S. 952	54.9	109.9	138.4	28.5	70.9	141.7	178.5	36.8	0.7	0.9	0.2	56.7	57.1	0.4
Blanes O. S. 1294	44.0	112.3	160.0	47.7	34.3	87.6	124.8	37.2	0.4	0.5	0.1	55.8	56.6	0.8
Blanes O. S. 1512	46.1	129.0	150.0	21.0	29.5	82.6	96.1	13.5	0.4	0.4	0.0	53.0	53.8	0.8

Table 2 | Mercury (Hg) concentrations, Hg accumulation rates (Hg-AR), Hg inventories, and silt content in sediment cores from the study area. Hg concentrations and Hg-AR are shown for 1900, the 1970s, and 2010; Hg inventories for the periods 1900–1970 and 1970–2010; and silt content for the 1970s and 2010. Differences (Δ) between 2010 and the 1970s are shown in bold italics. Sediment cores affected

by bottom trawling are highlighted in blue, whereas undisturbed cores are shown in red. Hg concentration and Hg-AR decreased between the 1970s and 2010 in trawled cores but increased in undisturbed cores. Hg inventories for the Palamós C. 744 core, which does not extend back to 1900, were extrapolated and should be considered upper-limit estimates. Hg inventories for the Foix C. 1373 core, which does not extend to 2010, represent minimum estimates.

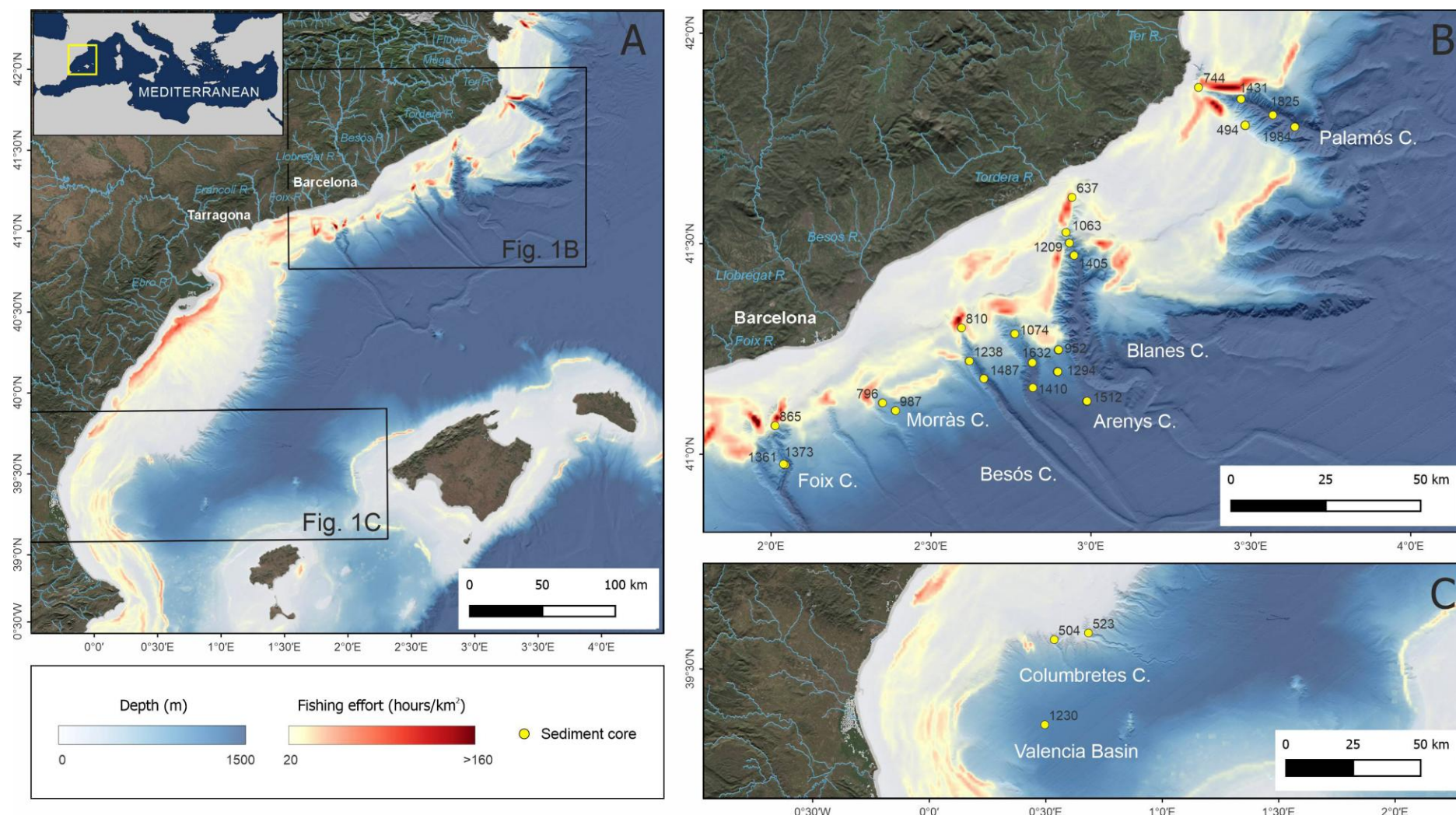


Fig. 1: Maps showing the location of the studied sediment cores. A general view of the study area in (A). Location of the sediment cores collected in the submarine canyons, canyon flanks and open slope of the northern and central Catalan margin (B) and cores taken in the canyons of the Valencia margin and in the Valencia Basin (C). Fishing effort is also represented to show the position of the fishing grounds. Note the higher values in the flanks of the submarine canyons incised in the northern and central and Catalan Margin.

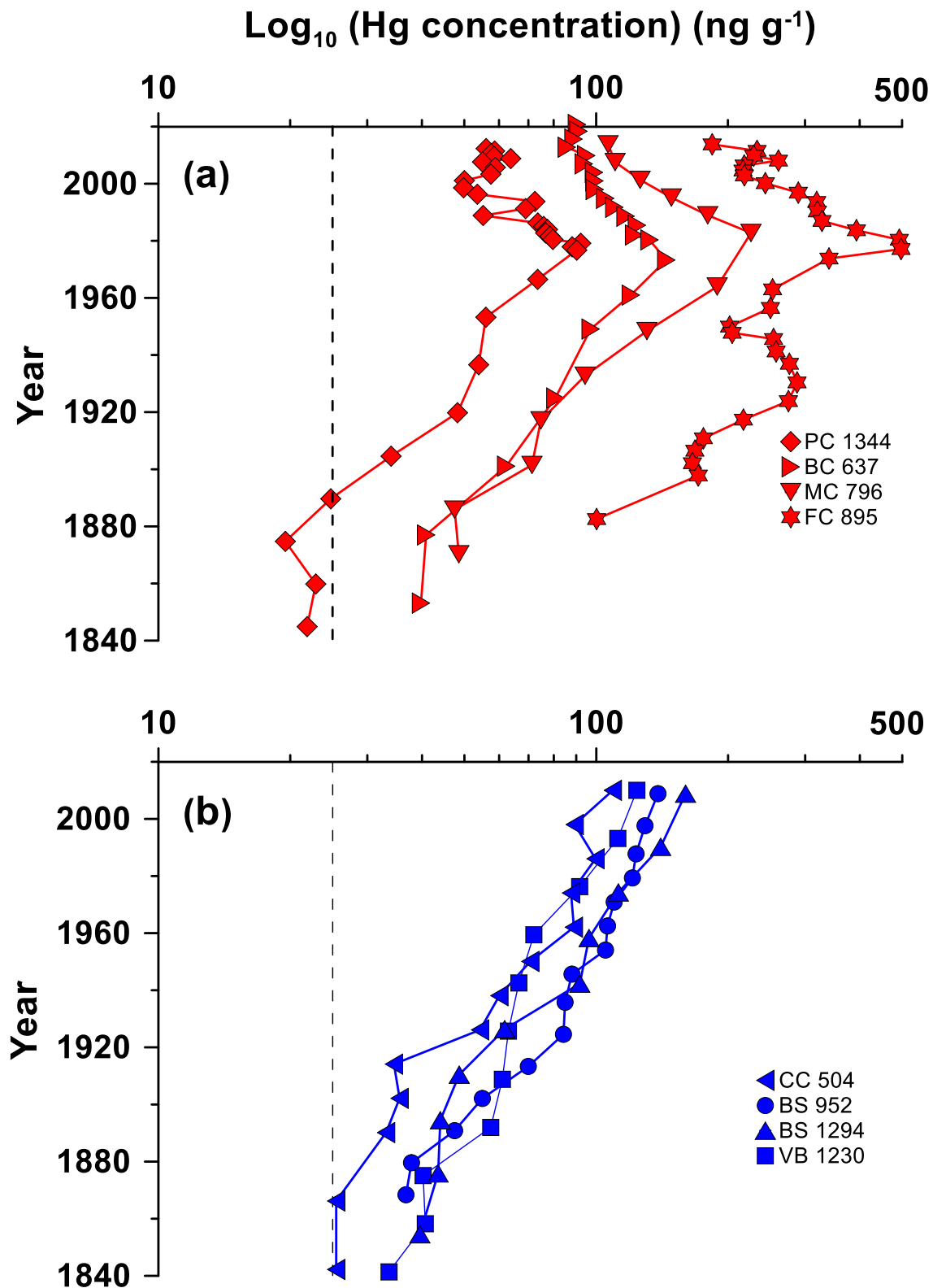


Fig. 2 | Vertical distribution of Hg concentrations through time in trawled and undisturbed marine environments. Vertical distribution of Hg concentrations versus age in sediment cores collected from selected submarine canyons affected by bottom trawling (a) and from selected sites unaffected by this activity (b). The black dashed line indicates the theoretical pre-industrial Hg baseline concentration of 25 ng g⁻¹ for the nineteenth century. PC, Palamós Canyon; BC, Blanes Canyon; MC, Morrás Canyon; FC, Foix Canyon; CC, Columbretes Canyons; BS, Blanes open slope; VB, Valencia Basin. Numbers in the legend indicate the water depth (m) of each sediment core.

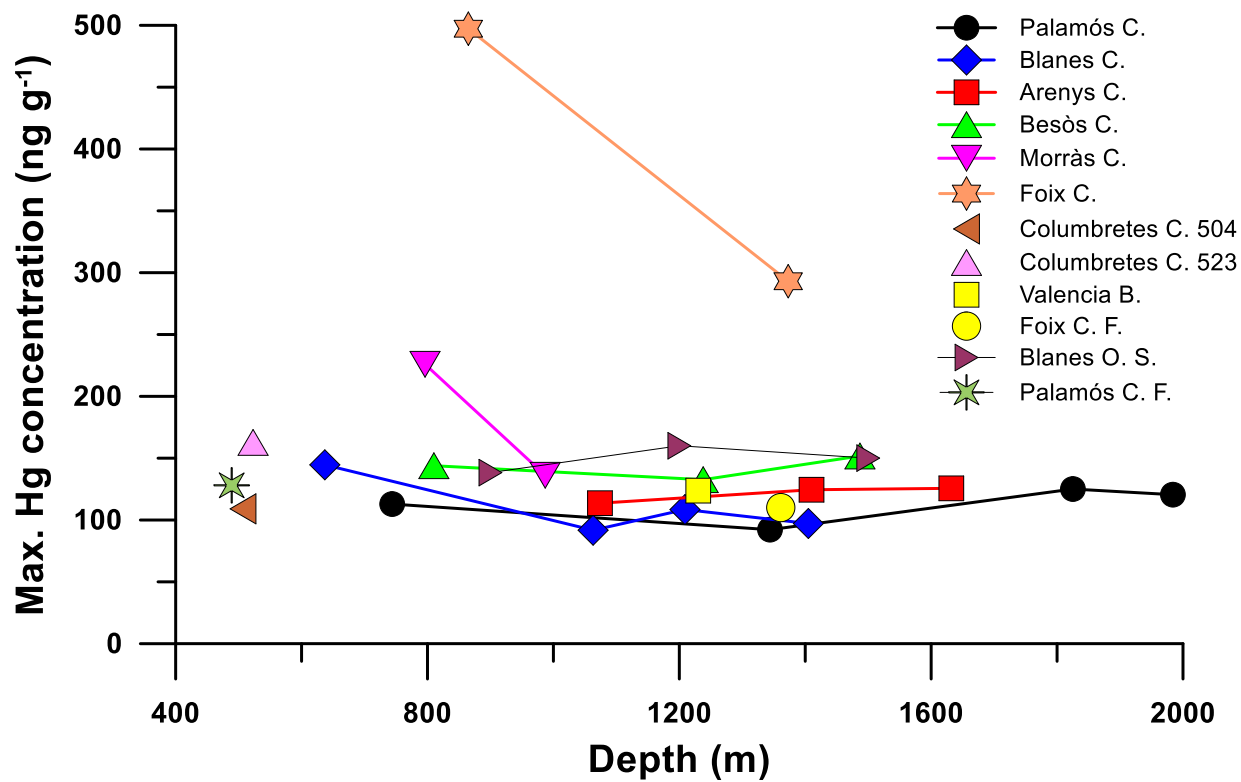


Fig. 3 | Maximum Hg concentrations versus water depth. Sediment cores are grouped according to depositional environment: submarine canyon (C.), canyon flank (C.F.), open slope (O.S.), and intra-slope basin (B.).

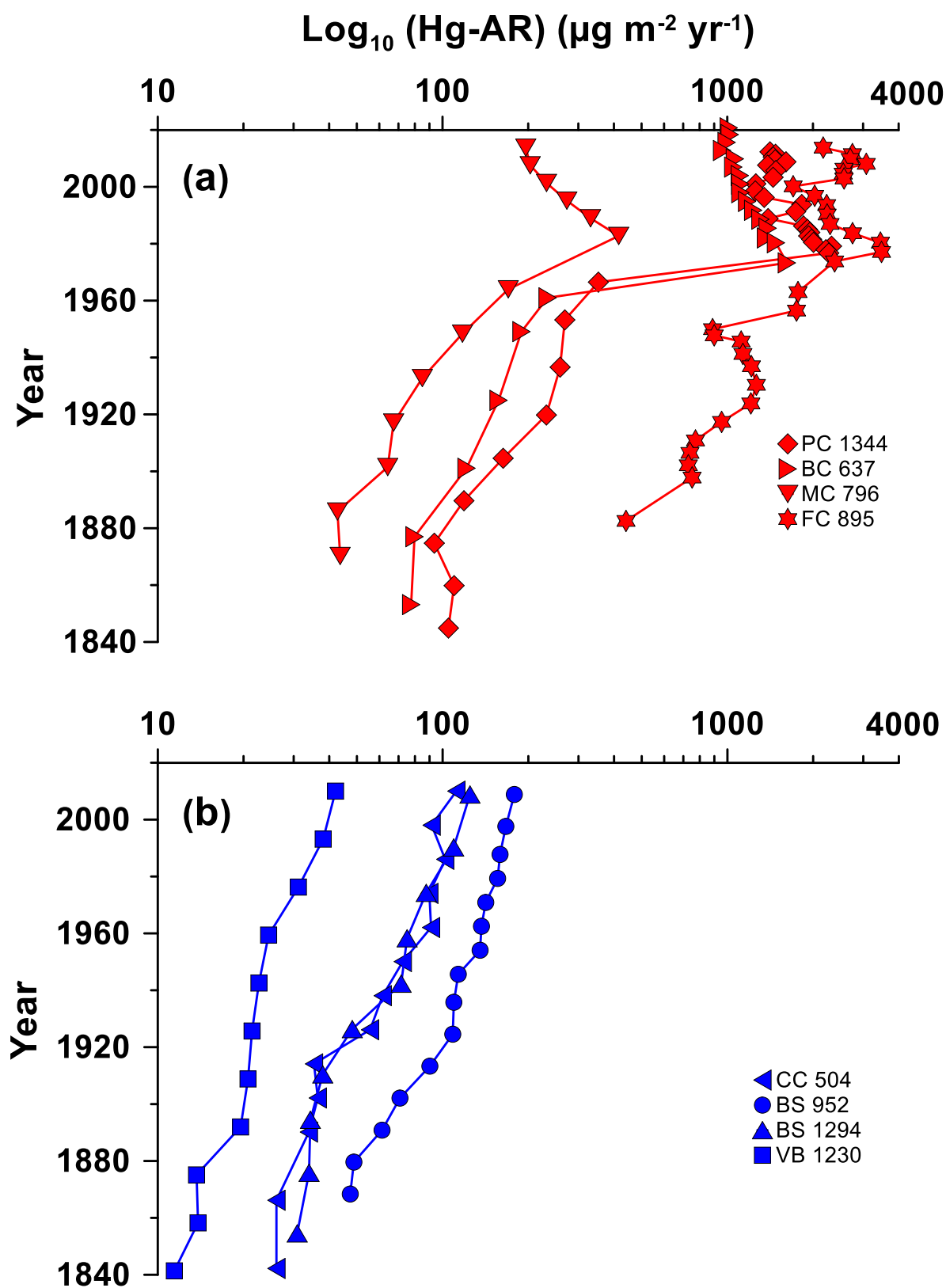


Fig. 4 | Contrasting Hg accumulation-rate histories in trawled and undisturbed marine environments. Vertical distribution of Hg accumulation rates (Hg-AR) versus age in sediment cores collected from selected submarine canyons affected by bottom trawling (a) and from selected sites unaffected by this activity (b). PC, Palamós Canyon; BC, Blanes Canyon; MC, Morràs Canyon; FC, Foix Canyon; CC, Columbretes Canyons; BS, Blanes open slope; and VB, Valencia Basin. Numbers in the legend indicate the water depth (m) of each sediment core.

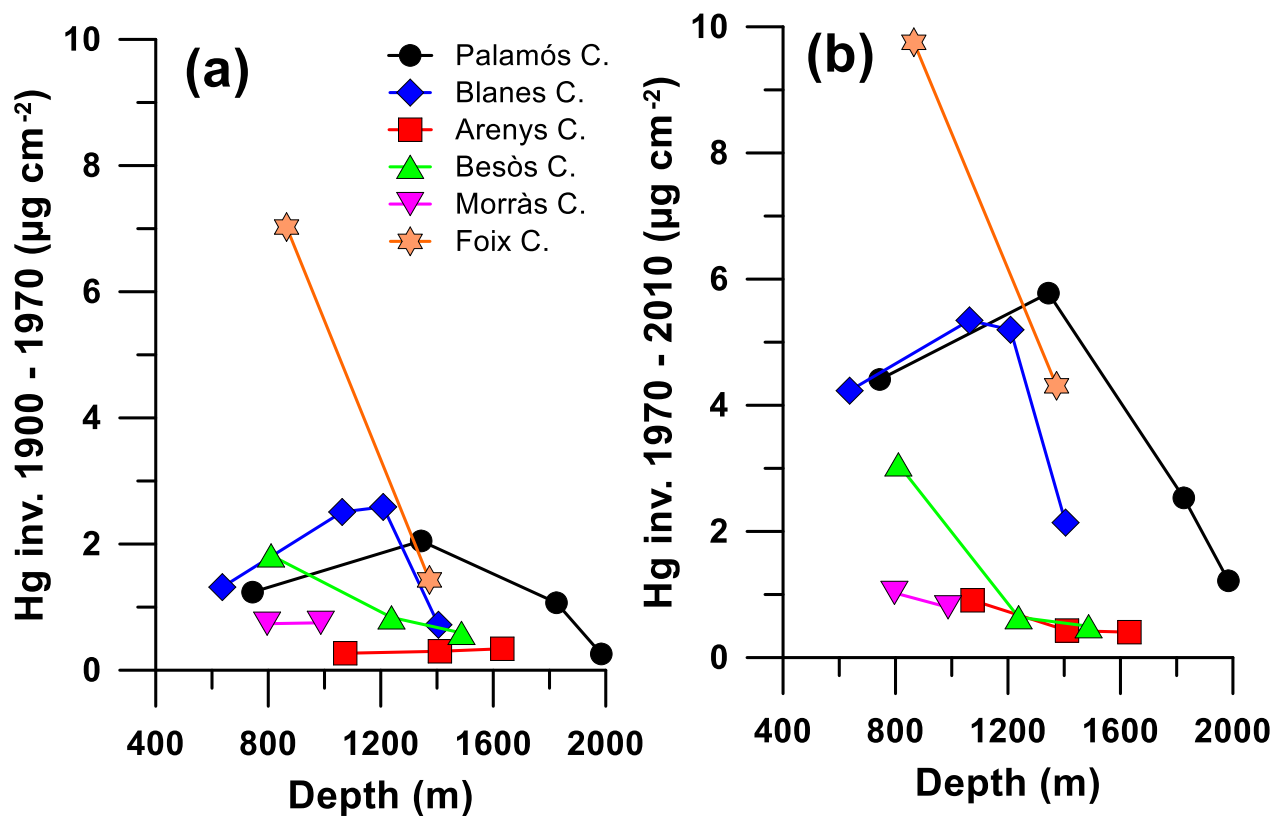
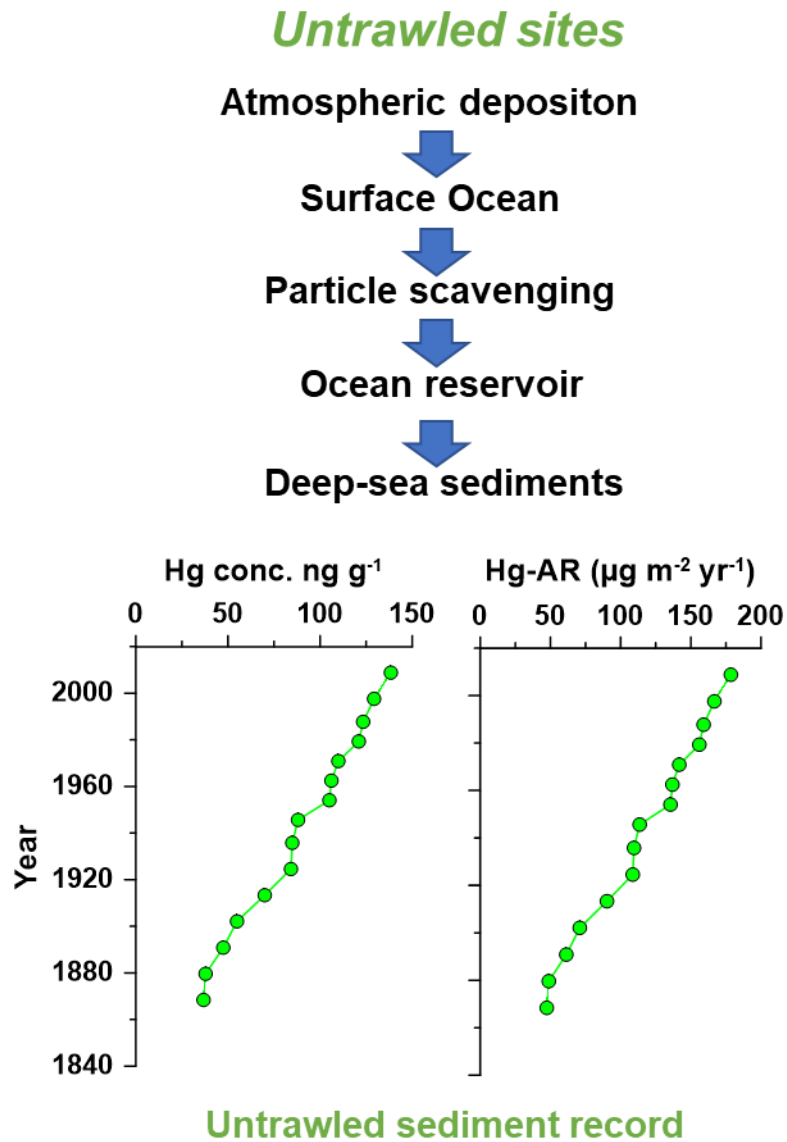
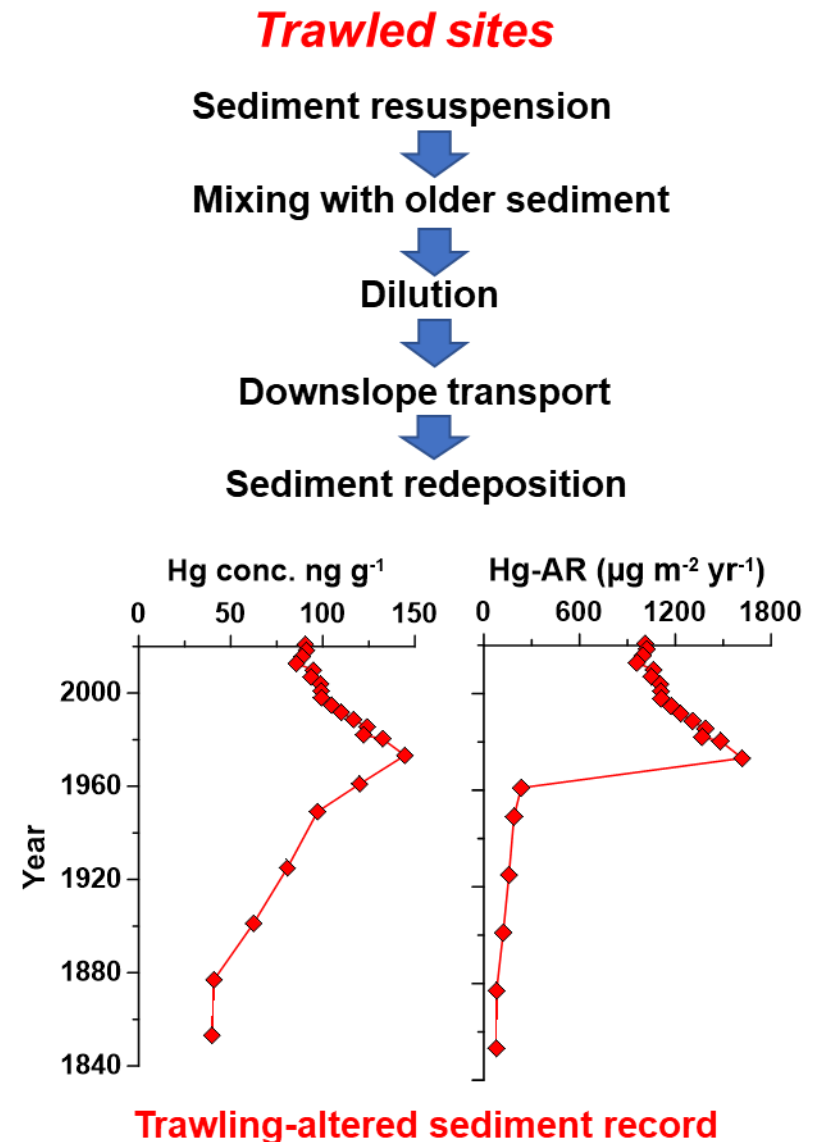


Fig. 5 | Water-depth distribution of Hg inventories in trawled submarine canyons before and after the 1970s. Hg inventories (Inv.) in trawled submarine canyons versus water depth for the periods 1900–1970 (a) and 1970–2010 (b). Hg inventories for the Palamós Canyon 744 m core, which does not extend back to 1900, were extrapolated in (a) and are therefore likely overestimated. Hg inventories for the Foix Canyon 1373 m core, which does not extend to 2010, are underestimated in (b). C., canyon.



Delayed response to changing Hg emissions



Enhanced Hg burial despite lower concentrations

Fig. 6. Conceptual model illustrating the contrasting controls on Hg records in deep-sea sediments. In untrawled environments, the large ocean Hg reservoir delays the response of deep-sea sediments to changes in atmospheric Hg emissions. In trawled submarine canyons, sediment resuspension, mixing with older sediment, dilution, downslope transport and redeposition overprint this natural signal, leading to enhanced Hg burial despite declining sediment Hg concentrations.

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