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Research Paper

Effects of bottom trawling on trace metal contamination of sediments along the submarine canyons of the Gulf of Palermo (southwestern Mediterranean)

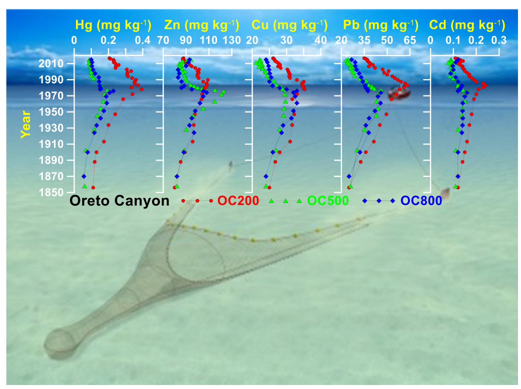
Albert Palanques [a,*], Sarah Paradis [b,c], Pere Puig [a], Pere Masqué [b,d,e], Claudio Lo Iacono [a]

[a] Institute of Marine Sciences, Consejo Superior de Investigaciones Científicas, Barcelona 08003, Spain
[b] Institute of Environmental Science and Technology (ICTA) and Physics Department, Universitat Autònoma de Barcelona, Bellaterra 08193, Spain
[c] Geological Institute, Department of Earth Sciences, ETH Zürich, 8092 Zürich, Switzerland
[d] International Atomic Energy Agency, 4a Quai Antoine 1er, 98000, Principality of Monaco, Monaco
[e] School of Natural Sciences, Centre for Marine Ecosystems Research, Edith Cowan University, Joondalup, WA 6027, Australia

HIGHLIGHTS

- Submarine canyons transfer trace metal contamination downslope.
- Submarine canyons can preserve the historical sediment record of trace metal contamination.
- Trace metal contamination in the sedimentary record increased until the 1970s and 1980s.
- Contamination decreased after the expansion of trawling fleets to deeper fishing grounds.
- Resuspension induced by bottom trawling decreased sediment trace metal contamination levels.

GRAPHICAL ABSTRACT



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ABSTRACT

Submarine canyons are preferential pathways for transport of particulate matter and contaminants from the shelf to the deep sea. The Gulf of Palermo continental margin has a very narrow shelf (about 2–3 km wide on average) and is incised by several submarine canyons that favour shelf-slope sediment transfer. A sediment core collected on the outer shelf and six sediment cores taken at different depths along the Oreto, Eleuterio and Anerella submarine canyons were analysed to study the transfer and historical record of trace metal contamination in the Gulf of Palermo continental margin. Trace metals, major elements, organic carbon and sediment grain size were analysed in these cores, which were dated with ^{210}Pb to assess their historical compositional evolution since the late 19th century. Hg, Pb, Cu, Zn and Cd content increased until the 1970s and 1980s, associated with the increase in urbanization and industrial activities in the Palermo area, and Hg was the contaminant that reached the highest enrichments. However, the increasing trend of these metals contamination was reversed in the 1970s and 1980s, coinciding with drastic changes in the terrigenous content and grain size of sediments in the canyon axes. These changes occurred when bottom trawling fleets expanded to deeper fishing grounds equipped with powerful trawlers around the Gulf of Palermo canyon heads and flanks and along the Oreto canyon axis. Bottom trawlers have resuspended large amounts of sediment, which have been transferred into the canyons since the 1970s and 1980s and have thus increased sediment accumulation rates. This resuspended sediment has been mixing with the sediment transferred and accumulated along the canyons, diluting and reducing its trace metal contamination levels since the expansion of the bottom trawling fleets.

* Corresponding author.
E-mail address: albertp@icm.csic.es (A. Palanques).

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1. Introduction

Trace metal (TM) contamination has been intensively studied in marine sediments for decades because they provide an integrative matrix reflecting the state of contamination in a given area (Bellas et al., 2011; Roberts, 2012). Fine sediment particles and the associated TMs have the same transport and deposition dynamics (Venkatesan et al., 1980; Salomons and Förstner, 1984; Fernex et al., 1986; Fimney and Huh, 1989; Garnier et al., 1991; Turner and Millward, 2002). Therefore, sediments can be a reservoir for contaminants and also a source of them when they are resuspended by natural processes such as bioturbation and wave-current interactions, or by anthropogenic activities such as dredging and trawling (Fichet et al., 1999; Eggleton and Thomas, 2004; Bradshaw et al., 2012; Rial and Beiras, 2012; Puig et al., 2012; Martín et al., 2014a; Palanques et al., 2020).

Many studies have focused on the spatial distribution and levels of sediment contamination in shallow marine areas close to sources of contamination, such as large cities and contaminated river mouths (e.g. Frignani et al., 1997; Cearreta et al., 2000; Bay et al., 2003; Dong et al., 2012). However, fewer have studied the dispersal of contaminated sediments into the deep sea, which has wider implications on assessing the impact of anthropogenic contamination through a complete source-to sink-system (Mil-Homens et al., 2013; Cossa et al., 2014). Therefore, knowledge of the distribution and fate of contaminants in marine sediments, particularly in deep areas, is still limited (Azaroff et al., 2020).

The continental slope can receive natural and anthropogenic TMs from direct atmospheric deposition onto the sea surface, from advection of riverine suspended particles and from resuspension of continental shelf sediments (Hickey et al., 1986; Monaco et al., 1999; Puig et al., 2014; Cossa et al., 2014).

In continental slope environments, submarine canyons can function as sediment traps and preferential conduits of particles from shallow to deep environments (Mullenbach and Nittrouer, 2000; Puig et al., 2003; de Stigter et al., 2007; Puig et al., 2014). Therefore, they can transfer and accumulate contaminated sediments discharged from the continent, especially if they are incised near urban and industrial development zones (Maurer et al., 1994; Puig et al., 1999; Hung and Hsu, 2004; Roussiez et al., 2005; Palanques et al., 2008; Costa et al., 2011; Salvado et al., 2012).

Although TM contamination in some submarine canyons is higher than that on the shelf (Maurer et al., 1994), most submarine canyons show lower contamination levels than shallow environments (Palanques et al., 2008; Hung et al., 2009). This is partly because submarine canyons act as corridors for sediment transport across the slope and distribute the particles coming from the shelf along their path (Emery, 1960; McHugh et al., 1992; Canals et al., 2006; Puig et al., 2008; Palanques et al., 2012; Martín et al., 2013). Indeed, multiple deposition and resuspension cycles can occur downcanyon (Palanques et al., 2012; Puig et al., 2014), leading to the dilution of TM contamination levels by mixing contaminated with uncontaminated particles, resulting into a general decrease in sediment contamination levels with depth.

Another anthropogenic process to consider in addition to natural sediment resuspension and transport processes is bottom trawling, which is a relevant activity affecting present-day sediment dynamics (Puig et al., 2012). In fact, bottom trawling has altered sedimentation processes on many continental shelves and submarine canyons for several decades. Trawling reworks and resuspends large volumes of sediment, which can be transferred to the deep sea as enhanced nepheloid layers (Martín et al., 2014b; Wilson et al., 2015; Arjona-Camas et al., 2019) or sediment gravity flows (Palanques et al., 2006; Martín et al., 2007; Puig et al., 2012; Martín et al., 2014c). However, the effects of the interactions between trawling and TM contamination on marine sediments are still poorly studied.

One of the most contaminated areas in the Mediterranean is the Gulf of Palermo (Fig. 1), which has experienced increased environmental degradation over the last 100 years (e.g., Di Leonardo et al., 2007, 2009; Tranchina et al., 2008; Caruso et al., 2011) owing to urban development and expansion of industrial activities. The lack or malfunctioning of water purification plants (Venezia, 1998; Caruso et al., 2011) and the usual discharge of untreated sewage and agricultural effluents has caused a steady decline of the water quality in the gulf (Di Leonardo et al., 2007). Because of this, in 2002 the Gulf of Palermo was included in the National Remediation Plan by the Italian Environmental Ministry (Di Leonardo et al., 2007), which aimed to increase the sustainable use of water resources. The gulf is heavily contaminated near the port of Palermo and around the adjacent Oreto River mouth (Tranchina et al., 2008; Caruso et al., 2011; Basile et al., 2011; Fig. 1). Offshore, contamination has been detected as far as the outer continental shelf but not on the continental slope, where only one core has been analysed on the lower slope (Di Leonardo et al., 2007, 2009, 2012).

Bottom trawling has been also affecting the bottom sediment of the Gulf of Palermo, resuspending and transferring sediments through the submarine canyons for decades and inducing drastic increases of the sedimentation rates (Paradis et al., 2021a), as has been observed in other submarine canyons (e.g. Martín et al., 2008; Puig et al., 2015; Paradis et al., 2018b),

In order to assess the role of submarine canyons incising the Gulf of Palermo as preferential conduits of sediments and their associated contaminants, previously dated sediment cores (Paradis et al., 2021a) collected on the shelf and along the axis of three submarine canyons (Fig. 1) were analysed to study the shelf-slope transfer and the historical evolution of TM contamination in this area.

The first aim of this paper is to study the characteristics and composition of the sediments within the Gulf of Palermo submarine canyons, assessing their levels of TM contamination from the late 19th century until present. The second aim is to discuss the potential factors controlling the historical evolution of sediment characteristics and TM contamination within the canyons and to explore the interactions between this contamination and bottom trawling.

2. Methods

2.1. Study area

The Gulf of Palermo is located in the northwest sector of the Sicilian continental shelf (Fig. 1) and extends for about 25 km alongshore in a west–east direction between the Gallo and Zafferano capes (Lo Iacono et al., 2011). The continental shelf of this gulf is very narrow (2–3 km wide on average), and its westernmost sector is the narrowest (Lo Iacono et al., 2011). The shelf edge, from 120 to 130 m depth, is incised by the heads of several submarine canyons, which extend downslope until a maximum depth of 1500 m (Lo Iacono et al., 2011, 2014) (Fig. 1). Three submarine canyons, the Arenella, Addaura and Mondello canyons, incise the steep slope of the western sector. Their evolution was mostly determined by upslope retrograding mass failures at their heads (Lo Iacono et al., 2011, 2014). The central and eastern sectors of the gulf are incised by the two largest submarine canyons, the Oreto and Eleuterio canyons. The Oreto Canyon has a regular V-shaped sinuous incision and its evolution is likely controlled by sediment input from the Oreto River, as suggested by the presence of several buried palaeovalleys in the shelf sector between the canyon head and the river mouth (Lo Iacono et al., 2011). The easternmost Eleuterio Canyon has a complex geomorphology, mostly controlled by mass wasting processes, which widen its head, breaching the shelf edge less than 1 km from Cape Zafferano (Lo Iacono et al., 2014).

Circulation on the northern Sicilian shelf is controlled by Modified Atlantic Water (MAW) that flows along shelf from west to east (Pinardi and Masetti, 2000; Caruso and Cosentino, 2008).

Two main rivers flow into the Gulf of Palermo, the Oreto and Eleuterio rivers, which flow through the city and receive the discharge from domestic and industrial sewages (Tranchina et al., 2008) (Fig. 1). Two minor seasonal rivers, the Kemonia and Papireto, were merged and canalized together with city sewage and currently flow as subterraneous channels into the old port of Palermo, named “La Cala”.

The primary sources of contaminants discharged into the Gulf of Palermo are urban and harbour activities (Di Leonardo et al., 2007, 2009; Tranchina et al., 2008; Basile et al., 2011). The Palermo urban area has about 900,000 inhabitants and hosts a variety of industrial and commercial

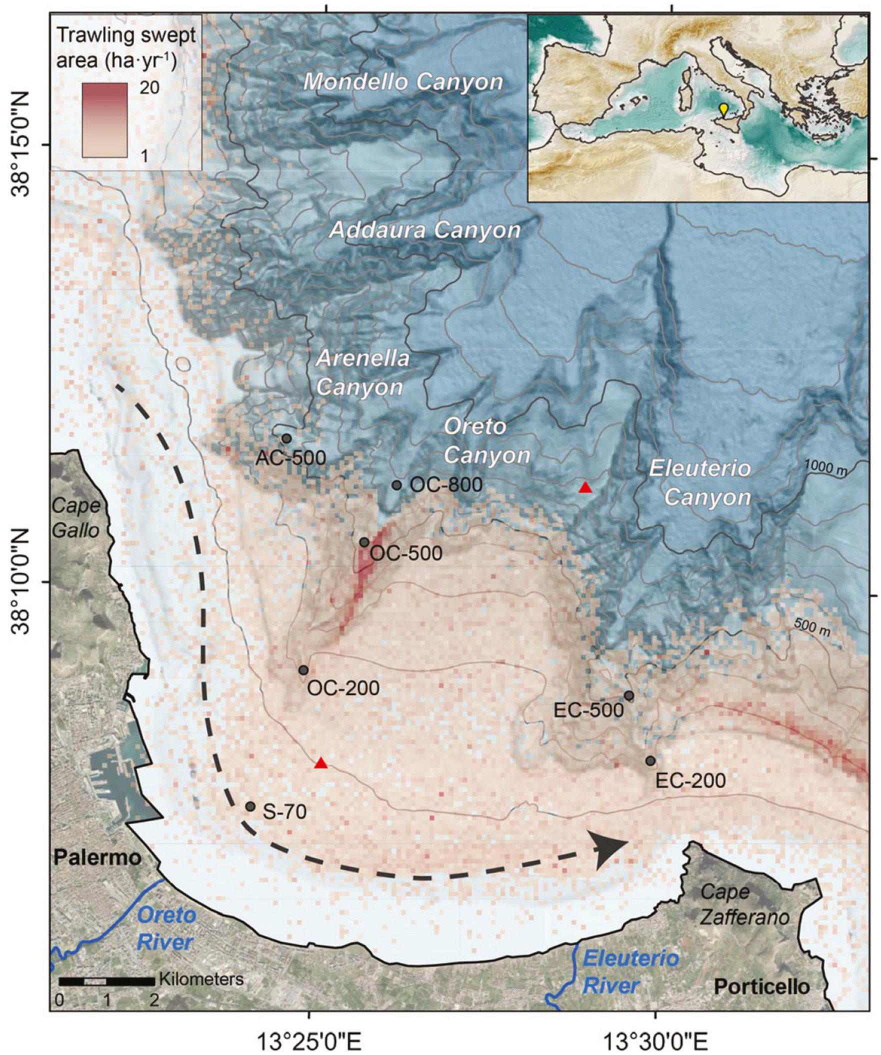

Fig. 1. Bathymetic map with 100 m contour interval, showing the Arenella, Oreto, and Eleuterio submarine canyons, the main rivers of the Gulf of Palermo, the main harbours (Palermo and Porticello) and the location of the sampled sediment cores in 2016 in black dots (•). The red triangles are the positions of the two cores taken in 2003 at 100 m and 700 m depth (Di Leonardo et al., 2007, 2009). Mean annual trawling intensity is given as swept area per year between 2008 and 2016 (from Paradis et al., 2021a). The dashed line shows the direction of the regional current and contour lines are displayed every 100 m.

activities. The port of Palermo, the largest in Sicily and one of the largest in Italy, has dockyards for building and repairing ships (Tranchina et al., 2008) and also receives the discharge of three pipelines with untreated city sewage, including the waste from several small goldsmiths containing mercury (Caruso et al., 2011).

Surface sediment collected on the inner shelf of the Gulf of Palermo is heavily contaminated with Hg, Zn, Cu, and Pb in the port area (2.7, 752, 698 and 220 mg kg^{-1}, respectively) and with Hg and Zn near the Oreto River mouth (1.1 and 220 mg kg^{-1}, respectively), while the TM levels are lower in the western sector of the gulf (Tranchina et al., 2008; Caruso et al., 2011; Basile et al., 2011). The TM contamination also reaches the outer shelf, where a dated sediment core taken at 100 m depth in 2003 showed an increasing Hg contamination over time up to 0.70 mg kg^{-1}, as well as moderate enrichments of Pb and Cu (Di Leonardo et al., 2007, 2009). On the open slope, a sediment core taken at 712 m depth near the Eleuterio Canyon, also in 2003, showed no significant contaminant signal (Di Leonardo et al., 2009, 2012). The locations of these two cores are represented by red triangles in Fig. 1.

The Gulf of Palermo also has some of the most important bottom trawling grounds in the region. Engine-propelled trawlers began to work in the gulf during the 1950s, but it was not until the mid-1960s and 1970s that the size of fishing fleets from the Palermo and Porticello ports grew. Nevertheless, they did not exceed 200 hp average engine power and operated mainly in shelf waters (Paradis et al., 2021a).

Since the late 1970s, the industrialization of bottom trawling has allowed these fleets to expand to deeper fishing grounds using more powerful trawlers (500–2000 hp). Bottom trawling became concentrated mainly on the outer shelf and upper slope sectors adjacent to the canyons of the gulf and on the Oreto Canyon axis. In this canyon, the greatest trawling effort occurs down to a depth of 700 m, whereas the axes of the other canyons are untrawled (Fig. 1). Concurrent with the expansion of fishing fleets to deeper fishing grounds in the late 1970s, sedimentation rates in the canyon axes increased by one order of magnitude (Paradis et al., 2021a).

2.2. Sampling

Seven sediment cores were collected in the Gulf of Palermo in 2016 with a K/C Denmark A/X six-tube multicore during the ExplorIng SiciLian CAnyoN Dynamics (ISLAND) cruise funded by the EU FP7 Eurofleets2 Project. Cores were sliced at 1 cm interval, and the slices stored in sealed plastic bags at −20 °C until they were freeze-dried in the laboratory for analysis. One sediment core was collected on the shelf at 70 m water depth (S-70), ~2.5 km off the Oreto River mouth and the Palermo harbour (Fig. 1). The other six cores were collected within three shelf-incised submarine canyons (from west to east: the Arenella, Oreto and Eleuterio canyons) at 200, 500 and 800 m. Three cores were taken in the axis of the Oreto Canyon (OC-200, OC-500, OC-800), two in the axis of the Eleuterio Canyon (EC-200, EC 500) and one in the axis of the Arenella Canyon (AC-500) (Fig. 1).

2.3. Analytical techniques

2.3.1. Grain size analysis and geochemical analysis

Grain size fractions were determined using a Horiba Partica LA950V2 particle size analyser. Prior to analysis, sediment aliquots of 1–4 g dry weight were oxidized using 20% H_2O_2. and disaggregated using 2.5% $P_2O_7^{4-}$. For geochemical analysis, the sediment samples were ground and homogenized in an agate mortar.

Total and organic carbon and total nitrogen contents were measured using a Thermo EA 1108 elemental organic analyser. Samples for organic carbon (OC) analysis were first decarbonated using repeated additions of 100 μL 25% HCl with 60 °C drying steps in between until no effervescence occurred. Carbonate content was calculated assuming all inorganic carbon is contained within the calcium carbonate ($CaCO_3$) fraction, thus using the molecular mass ratio 100/12.

For the analysis of major and minor elements and TMs, a total digestion technique was carried out according to Querol et al. (1996). It consists of: a) digestion of volatile elements from 0.1 g of sediment sample in a closed system with 2.5 mL of concentrated nitric acid MERK supra-pure at 90 °C for 2 h; and b) digestion of non-volatile elements with 7 mL of supra-pure hydrofluoric acid and heating at 90 °C in a closed bomb for 3 h and the addition of 2.5 mL of supra-pure perchloric acid and 2.5 mL of nitric acid. When completely dry, 2.5 mL of supra-pure nitric acid was added to the sample and the solution was transferred to a metered flask.

For every 18 samples, a blank, a PACS-2 reference material (National Research Council, Canada) and a random-replicated sample were used for analytical quality control. Zn, Cu, Pb, Cd, Ni, Cr, Mn and Li were analysed by ICP-MS and Al, Fe and Ca were analysed by ICP-OES. The overall analytical uncertainty was below 15% and typically between 5% and 10%.

For Hg analysis, a LECO AMA254 Mercury Analyser complying with US EPA Method 7473 (US EPA, 2007) was used. 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%.

Normalizing trace elements to Li and Al contents led to very similar results and interpretations concerning the temporal evolution of anthropogenic contamination. Lithium was chosen as normalizer element to compensate for grain-size variability, to take into account the clay fraction, and to identify anomalous concentrations of heavy metals in sediments. Li is incorporated in fine-grained aluminosilicate metal-bearing minerals, but not in the Al-rich but metal-poor feldspars that occur throughout the grain size spectrum of such sediments (Loring,1990). Similar normalization method was already used successfully for Mediterranean sediments (Aloupi and Angelidis, 2001; Cossa et al., 2014) and for Atlantic sediments (Costa et al., 2011). Enrichments Factors (EF) were calculated using the equation:

$$EF = (M/Li)_{sample}/(M/Li)_{background}$$

where M is the element concentration. Background TMs were obtained from bottom sediment sections of the canyon sediment cores dated before 1870 assuming constant sedimentation during that period. An EF greater than 2 indicates a large proportion of metal delivered from non-crustal material, which is from anthropogenic, biogenic and/or diagenetic sources (Sutherland, 2000).

For the assessment of toxicity risks, we used the reference values described by Long et al. (1995), which are also used by the National Oceanic and Atmospheric Administration (NOAA) as a reference to assess toxicity. These authors define two values for each TM: the effect range low (ERL) and the effect range median (ERM). This allowed us to establish three ranges according to which the effects on the environment are predicted to be minimal (value < ERL), occasional (ERL < value < ERM) or frequent (value > ERM) (Table S1). However, these guidelines are only an approach that must be considered carefully as they do not take into account the chemical forms in which the elements are in the sediments, the grain-size effect and the regional biotic and environmental variability. In addition, for Hg there was relatively weak relationships between its concentrations and the incidence of effects (Long et al., 1995). This is why we use the term “contamination”.

2.3.2. Statistical analysis

Principal Components Analysis (PCA) was applied to examine the different groupings and their geochemical and source characteristics and to reduce the complexity of the original data in the study area. Geometrically, this new set of variables represents a principal axis rotation of the original coordinate axes of the variables around their mean (Jackson, 2003; Huang et al., 2010). The factor analysis was performed by using the statistical software IBM SPSS Statistics version 27.

2.3.3. Sediment age models

The average sedimentation rates at each site were obtained from excess ^{210}Pb concentration profiles using the CF:CS (constant flux: constant sedimentation, Krishnaswamy et al., 1971) model, as described in Paradis et al. (2021a). Variations in the slope of excess ^{210}Pb concentrations were attributed to either surface sediment mixing or changes in sedimentation rates (Paradis et al., 2017, 2018a and b). Since sediment mixing can also affect downcore excess ^{210}Pb concentrations, average sedimentation rates in cores with surface mixed layers were regarded as upper estimates (Nittrouer et al., 1979).

Section depth dates of each core were inferred on the basis of mass accumulation rates ($g \cdot cm^{-2} \cdot yr^{-1}$) and cumulative dry mass ($g \cdot cm^{-2}$) to correct for sediment compaction with core depth, and the associated errors were obtained by error propagation of the model outcomes. Surface mixed layers cannot be dated, and sections with constant excess ^{210}Pb concentrations, interpreted as the instantaneous arrival of allochthonous sediment, were given the same date as the overlying sections (Paradis et al., 2017, 2018a and b). In some cases, dates were extrapolated further downcore, beyond the depth limit of excess ^{210}Pb, assuming constant sedimentation rates below.

The sedimentation rates of the sediment cores were described and presented in detail in Paradis et al. (2021a). The sediment core collected on the continental shelf (S-70) had a single average sedimentation rate of 0.20 cm yr^{-1} with a 4-cm-thick surface mixed layer. In contrast, most of the cores taken in the submarine canyons axes showed a drastic change in the sedimentation rates, with basal values ranging between 0.11 and 0.16 cm yr^{-1} and increased sedimentation rates reaching up to 1.38 cm yr^{-1} occurring synchronously since the 1980s (Table 1). In the Oreto Canyon, the sedimentation rates increased by a factor of 3 at 200 m depth, by a factor of 5 at 500 m depth and by a factor of 2 at 800 m depth. (Table 1). In the core EC200 from the Eleuterio Canyon, the horizon where excess ^{210}Pb (supported ^{210}Pb concentrations) is no longer detected, was not reached because it did not sample sediments older than 1950, but the magnitude of its sedimentation rate since 1980 is similar to those of the other canyon cores.

Table 1
Sediment accumulation rates (SAR) before and after the 1980s of the sediment cores collected on the shelf (S-70) and in the Oreto Canyon (OC-200, OC-500, OC-800), the Eleuterio Canyon (EC-200, EC-500) and the Arenella Canyon (AC-500). SAR in EC 200 before 1980 could not be estimated. Data extracted from Paradis et al. (2021a).

Sediment core	SAR before 1980s ($cm\ yr^{-1}$)	SAR after 1980s ($cm\ yr^{-1}$)
S-70	0.203 ± 0.009	0.203 ± 0.009
OC-200	0.157 ± 0.009	0.59 ± 0,03
OC-500	0.114 ± 0.007	0.73 ± 0.05
OC-800	0.110 ± 0.006	0.210 ± 0.012
EC-200	–	0.52 ± 0.02
EC-500	0.114 ± 0.006	1.38 ± 0.07
AC-500	0.131 ± 0.007	1.37 ± 0.07

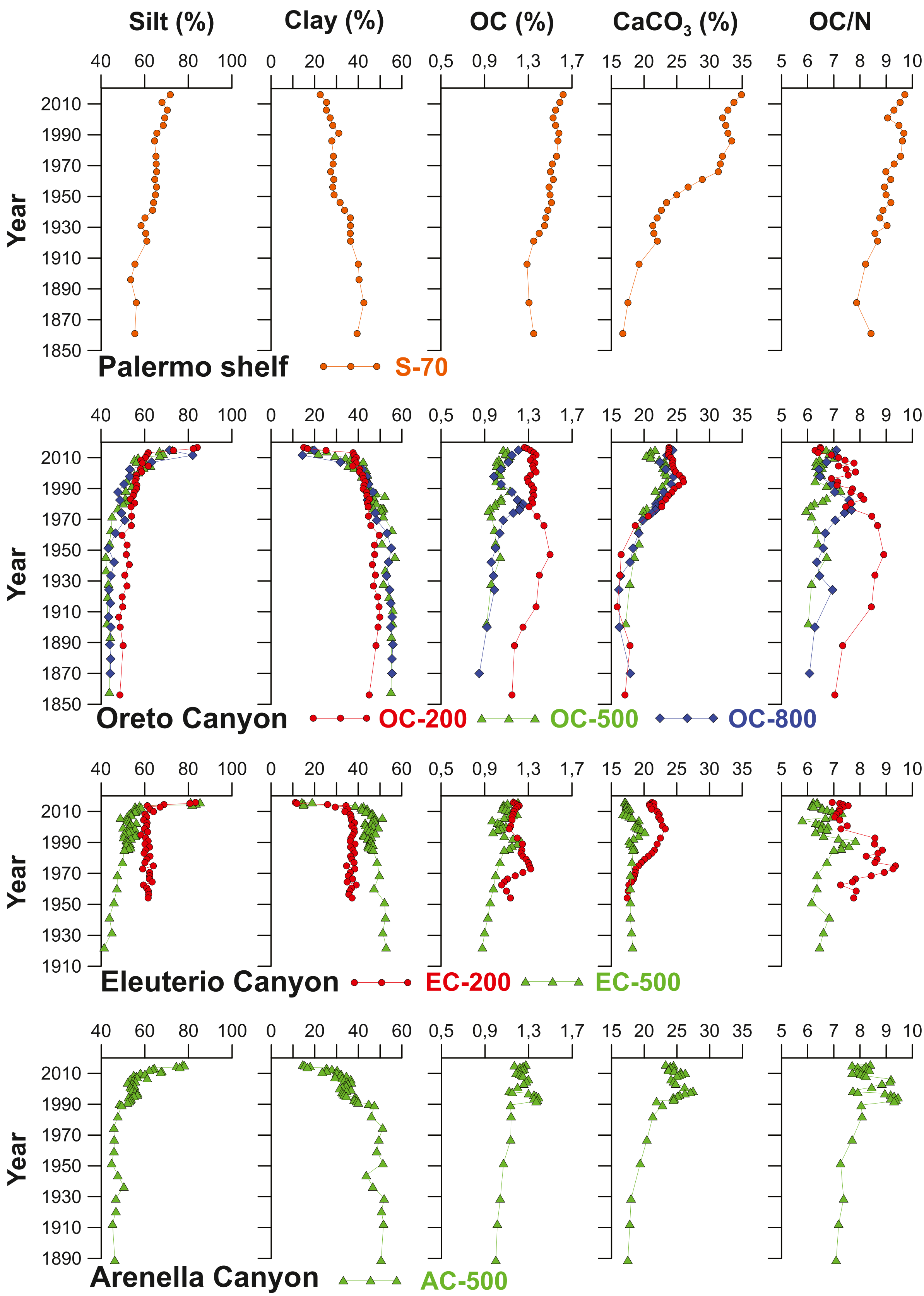

Fig. 2. Vertical distributions of silt, clay, organic carbon (OC), calcium carbonate ($CaCO_3$), and OC/N (mol:mol) versus age of sediment cores collected on the shelf (S70) and in the Oreto Canyon (OC-200, OC-500, OC-800), the Eleuterio Canyon (EC-200, EC-500) and the Arenella Canyon (AC-500).

The EC-500 core showed an increase in the sedimentation rate by one order of magnitude since the 1980s (Table 1). Similarly, in the Arenella Canyon, the AC-500 core also showed an increase by one order of magnitude since the 1980s (Table 1).

The results of all sedimentological and geochemical analyses discussed in the paper are presented versus time derived from the age models to study the trends of sediment and TM accumulation from the onset of the industrial age until present.

3. Results

3.1. Grain size, calcium carbonate, organic carbon and major elements

All the collected sediment samples consisted of fine sediments containing more than 90% of mud (<63 μm). However, all the studied cores showed an upwards trend of increasing silt content after the 1970s and 1980s, mirrored by a decreasing trend for clay (Fig. 2).

On the shelf, OC, $CaCO_3$ and OC/N increased upwards with some small variability after the 1970s (Fig. 2). In the canyon cores, OC and OC/N also increased upwards until about the late 1980s, after which they showed no clear trends, with both increasing and decreasing values. In both cores collected at 200 m water depth, OC and C/N tended to decrease after the 1970s, whereas in the deeper cores they increased between the 1970s and the 1980s, decreased during the 1990s and again increased from then on (Fig. 2).

The Ca content of the studied sediment cores showed an upward increasing trend against the decreasing trend of Li, Al and Fe. In the shelf sediment, these trends were more pronounced prior to the 1970s and 1980s, and the values became relatively constant afterwards (Fig. 3). In most of the canyon cores, the upward trend of increasing Ca and decreasing Li, Al and Fe became more pronounced from the 1970s and 1980s, changed to an opposite trend in the 1990s and changed again to either increasing or decresing trends from the 2000s (Fig. 3).

Mn showed a decreasing trend in the shelf core with a slight increase towards the surface. In the canyon cores, Mn was relatively constant with a small increase in the late 1980s in the cores taken at 500 m and 800 m depth, and increased drastically reaching maximum values towards the surface of all the cores (Fig. 3).

3.2. Trace metals

The sediment core taken on the shelf showed initially only a minor increase in TMs from the 1860s onward, followed by a more drastic increase from the 1910s and 1920s, more constant values from the 1930s to the 1960s and a slight decrease from the 1960s (Fig. 4). The shelf sediment had anthropogenic enrichments of Hg, Zn, Cu, Pb, and Cd (Table 3). Hg showed the highest contamination levels, with EFs of up to 32, followed by Pb and Cd with EFs of 5.6 and 4.9 respectively. Ni and Cr showed no significant anthropogenic enrichments (EFs <1.3). Mean and maximum Hg concentrations in the shelf core were above the ERM values and all Pb and Cu concentrations were above the ERL value (Table 2; Fig. 4).

The sediment cores from the submarine canyons showed an upward-increasing trend of Hg, Zn, Cu, Pb and Cd from around the 1910s to the 1970s and 1980s, when they reached maximum values and EFs (Fig. 4). From the 1970s and 1980s to the sampling date, there was a decreasing trend of these TMs in all the canyon cores.

Maximum Hg and Pb concentrations of the canyon cores were above the ERL values and reached EFs of up to 10.9 and 2.8, respectively (Table 2; Fig. 4). Maximum Cu concentrations were above the ERL value in some canyon cores, and the EFs of Zn, Cu and Cd were low (<2) but still perceptible. The highest Hg, Pb and Cd concentrations and EFs were in the Oreto and Eleuterio canyon heads (OC-200 and EC-200), whereas the maximum Zn values were in the Oreto and Eleuterio mid-canyon cores (OC-500 and EC-500). Cu values of the cores along the Oreto and Eleuterio canyons were similar among them (Fig. 4; Tables 2 and 3). Cr and Ni had natural values similar to those in the shelf core (Table 3).

3.3. Principal component analysis

Correlation coefficients between the analysed variables are shown in Table S2 and the results of the PCA in Fig. 5.

Component 1 accounted for 42.1% of the variance. It grouped Li, Cr, Ni, Fe, Al and clay with a positive contribution indicating that Cr and Ni were mainly associated with the fine-grained aluminosilicate clay minerals. The EFs of these TMs were no higher than 1.3, indicating that the sediments from the canyons were not significantly affected by anthropogenic Ni and Cr contamination. Component 1 also grouped Ca, sand, silt, $CaCO_3$, OC and C/N, but with a negative contribution, indicating that Ca and $CaCO_3$ were probably associated with biogenic carbonate in the sand and silt fraction and with more degraded organic matter. Therefore, component 1 represented the terrigenous elements against the biogenic carbonate elements (Fig. 5; Table S3).

Component 2 accounted for 20.7% of the variance. It grouped Cu, Zn, Cd, Pb, Hg, OC and OC/N with a positive contribution, thus including the TMs, which were not correlated with the terrigenous elements (Al, Li) and the clay fraction. These TMs showed moderate or even high EFs in the case of Hg. This component represented the TMs affected by anthropogenic contamination (Fig. 5; Table S3).

Component 3 accounted for only 9.9% of the variance. It receives the positive contribution of Mn, N and silt and the negative contribution of clay, and could represent early diagenesis effects in recently winnowed sediment (Table S3).

4. Discussion

4.1. Increase in anthropogenic TM contamination and transfer through the canyons

The presence of anthropogenic TMs in the environment has changed in the course of history, but although anthropogenic impacts started during Greek and Roman times, the highest TM levels have been reached mostly since the industrial revolution (Nriagu, 1996). Contamination increased substantially during the 20th century, associated with the boost of economical and industrial development generating a continuous enrichment of TMs in marine bottom sediments of many parts of the world (Palanques et al., 1998; Hung et al., 2009; Natesan and Seshan, 2010; Costa et al., 2011; Pan and Wang, 2011; Mil-Homens et al., 2013; Heimbürger et al., 2012; Cossa et al., 2014; Tamburrino et al., 2019).

Previous studies in the Gulf of Palermo showed that untreated sewage from the industrial, urban and port activities and the discharge of agricultural effluents contaminated especially the inner shelf sediment (Tranchina et al., 2008; Caruso et al., 2011), whereas their effects extended in lesser degree to the outer shelf (Di Leonardo et al., 2007, 2009, 2012).

On the inner shelf, the surface TM enrichments reached maximum values around the Palermo Port and the Oreto River mouth, decreasing sharply towards the west and more gradually towards the east (Tranchina et al., 2008; Caruso et al., 2011). The historical evolution of TM contamination in dated sediment cores taken in 2003 showed an increase in Hg and Pb from the 1920s on the outer continental shelf at 100 m depth, whereas there was no TM enrichment on the open continental slope at 700 m next to the Eleuterio Canyon (location in Fig. 1) (Di Leonardo et al., 2007, 2009, 2012). From these results it seemed that the TM contamination from the city and port of Palermo and the Oreto River was dispersed along and across the shelf without reaching the continental slope.

In our study, we recorded higher TM contamination levels in the outer shelf core (S-70) than those recorded previously on the outer shelf, and more importantly, we recorded for the first time anthropogenic TM enrichments in sediments from the continental slope in the submarine canyon axes mainly since the 1910s – 1920s (Fig. 4). The TMs enrichments in the canyons were lower than those of the shelf and decreased with depth (Tables 3 and 4), indicating downcanyon dilution due to sediment resuspension and transport processes occurring within these morphological features (Palanques et al., 2008; Hung et al., 2009). The canyon sediment cores taken at similar depths

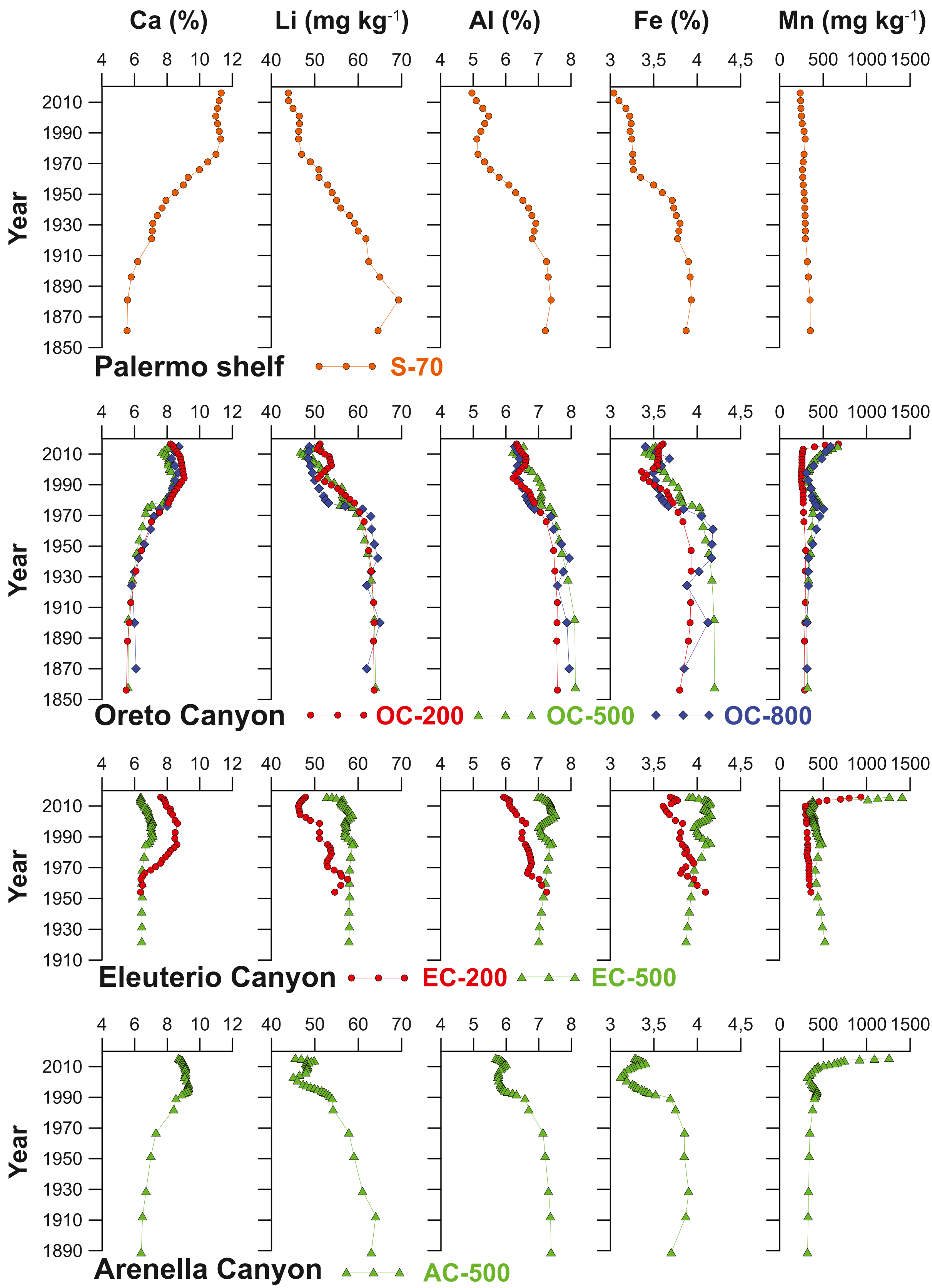

Fig. 3. Vertical distributions of Ca, Li, Al, Fe, and Mn versus age of sediment cores collected on the shelf (S-70) and in the Oreto Canyon (OC-200, OC-500, OC-800), the Eleuterio Canyon (EC-200, EC-500) and the Arenella Canyon (AC-500).

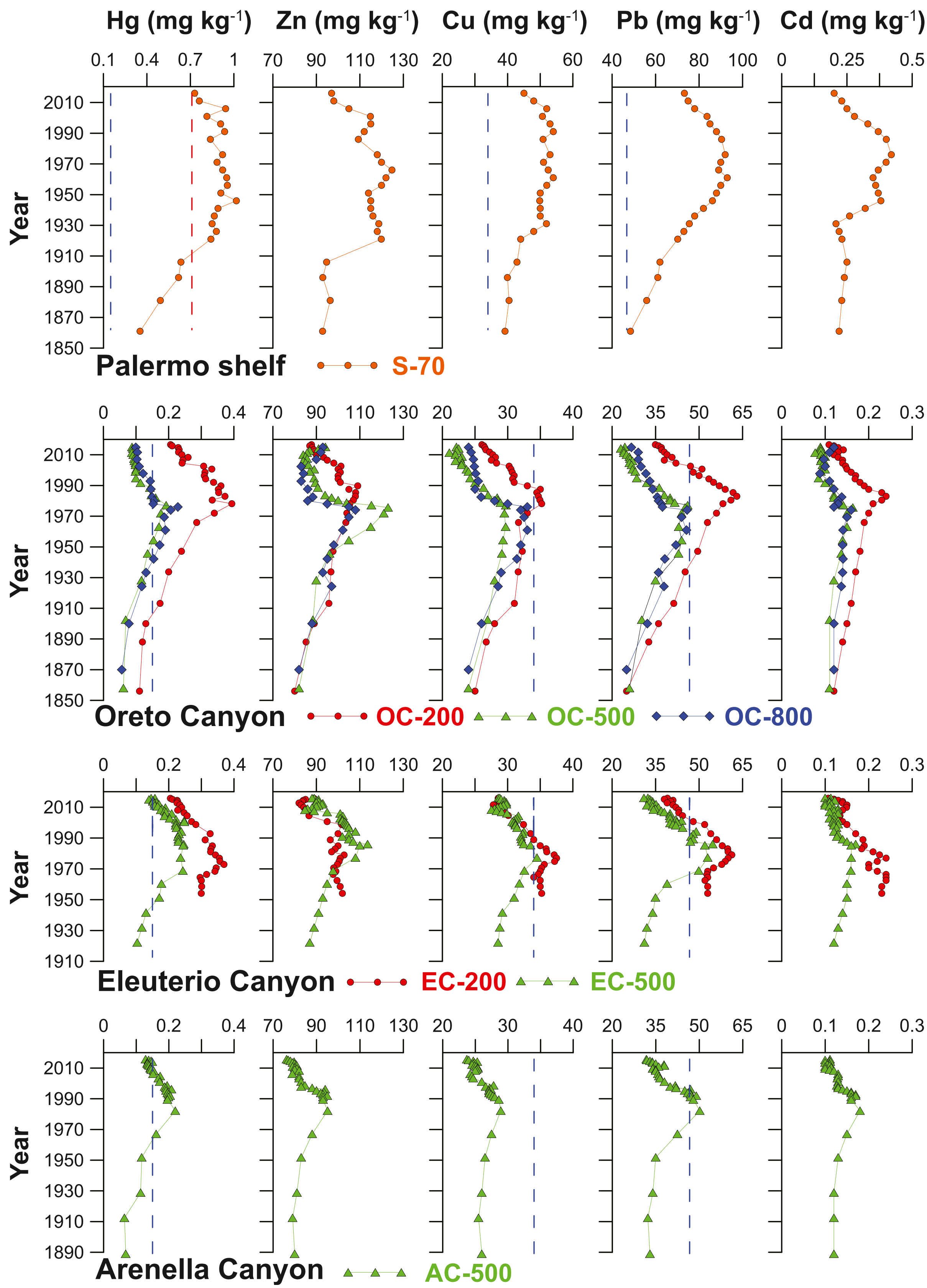

Fig. 4. Vertical distributions of anthropogenically enriched trace metals (Hg, Zn, Cu, Pb, Cd) versus age of sediment cores collected on the shelf (S-70) and in the Oreto Canyon (OC-200, OC-500, OC-800), the Eleuterio Canyon (EC-200, EC-500) and the Arenella Canyon (AC-500). Note different scale of the x-axis for the S-70 core. Blue dashed line: effect range low (ERL) value. Red dashed line: effect range median (ERM) value. Zn and Cd concentrations do not reach the ERL value.

Table 2
Minimum, maximum and mean concentrations of the enriched trace metals in the sediment cores collected on the shelf (S-70) and in the Oreto Canyon (OC-200, OC-500, OC-800), the Eleuterio Canyon (EC-200, EC-500) and the Arenella Canyon (AC-500). Location of cores shown in Fig. 1. Concentrations between ERL (effect range low) and ERM (effect range median) in blue, above ERM in red. ERL and ERM values from Long et al. (1995) are represented in supplementary Table S1.

Sediment Core		Hg (mg kg^{-1})	Zn (mg kg^{-1})	Cu (mg kg^{-1})	Pb (mg kg^{-1})	Cd (mg kg^{-1})	Al (%)	Li (mg kg^{-1})
S-70	Min-Max	0.33-1.02	88-125	37-54	56-93	0.20-0.42	4.9-7.4	44-69
	Mean	*0.80*	*109*	*48*	*77*	*0.29*	*6.0*	*53*
OC-200	Min-Max	0.11-0.39	78-109	23-35	21-63	0.11-0.24	6.2-7.6	50-64
	Mean	*0,26*	*96*	*30*	*46*	*0.16*	*6.8*	*56*
OC-500	Min-Max	0.06-0.20	77-123	21-30	23-47	0.08-0.16	6.2-8.1	46-64
	Mean	*0.13*	*96*	*26*	*34*	*0.11*	*7.1*	*56*
OC-800	Min-Max	0.04-0.23	78-108	22-34	19-46	0.09-0.16	6.3-7.9	48-65
	Mean	*0.14*	*93*	*28*	*35*	*0,13*	*7.0*	*56*
EC-200	Min-Max	0.21-0.37	82-104	28-37	38-62	0.11-0.24	5.9-7.3	46.3-58
	Mean	*0.29*	*93*	*32*	*50*	*0.17*	*6.6*	*51*
EC-500	Min-Max	0.1-0.25	85-114	28-35	31-55	0.10-0.17	7.0-7.5	53-59
	Mean	*0.19*	*97*	*30*	*40*	*0.13*	*7.2*	*57*
AC-500	Min-Max	0.06-0.22	76-95	24-29	32-50	0.1-0.18	5.7-7.4	45-64
	Mean	*0.16*	*84*	*26*	*39*	*0.13*	*6,1*	*51*

(200 and 500 m) showed similar trends of increasing TM contents and mean and maximum concentrations and EFs (Fig. 4 and Table 2). The maximum TM concentrations occurred mainly in the 1970s and 1980s and decreased from the 200 m to 500 m depth cores, except for Zn, maintaining similar values from the 500 m depth cores to the 800 m depth core.

All this indicates that the Gulf of Palermo submarine canyons transfer and accumulate along them TM-contaminated sediments originating from the adjacent shelf. The absence of TM enrichments in the core taken on the open slope (Di Leonardo et al., 2007, 2009, 2012) (location in Fig. 1) could suggest that submarine canyons are the main downslope conduits of TM-contaminated sediment in this continental margin (Fig. 4).

The concentration of Hg in the sediments of the Oreto, Eleuterio and Arenella canyons is similar to those reported for the Kaoping (Taiwan) and Cascais (Portugal) canyons (Hung et al., 2009; Mil-Homens et al., 2013), but lower than those of the Capbreton Canyon (France) (Azaroff et al., 2020).The concentrations of Cd, Cu, Pb and Zn measured in the Gulf of Palermo submarine canyons are similar to those reported for the Cap de Creus (Spain) and Capbreton canyons (Cossa et al., 2014; Azaroff et al., 2020) and slightly lower than those of the Foix Canyon (Spain) (Palanques et al., 2008).

The upward increase in the anthropogenic TM enrichments since the 1910s–1920s coincided quite well with the upward decrease in the fine-grained terrigenous fraction (Al, Li, Fe, and clay) in parallel with the upward increase in the coarse biogenic carbonate fraction (Ca, sand, silt, CaCO3 and OC) (Figs. 2, 3 and 4). These opposed trends were also observed in sediment cores from the Cascais Canyon and interpreted as a decrease in the fine-grained terrigenous inputs caused by the construction of dams and the decrease in the water discharge due to water consumption and climate change (Mil-Homens et al., 2021). In line with the above observations, the decrease in yearly precipitation and the increase of water consumption by agricultural, urban and industrial activities in the Gulf of Palermo has led to a decrease in the water discharge of the Oreto and Eleuterio Rivers from the early 20th century (Cannarozzo et al., 2006; Arnone et al., 2013; Billi and Fazzini, 2017) and therefore a decrease in their discharge of terrigenous sediments into the gulf since the 1910s and 1920s.

4.2. Decline of anthropogenic TM enrichments in the Gulf of Palermo

Following several decades of increasing contamination in marine sediments, technological improvements in the treatment of industrial and

Table 3
Minimum, maximum and mean enrichment factors of the trace metals in the sediment cores collected on the shelf (S-70) and in the Oreto Canyon (OC-200, OC-500, OC-800), the Eleuterio Canyon (EC-200, EC-500) and the Arenella Canyon (AC-500). Location in Fig. 1.

Sediment Core		Hg	Zn	Cu	Pb	Cd	Cr	Ni
S-70	Min-Max	8.5–32.6	1.1–2.0	1.5–2.9	2.1–5.6	1.8–4.9	1.0–1.3	1.0–1.1
	Mean	*25,0*	*1.7*	*2.4*	*4.4*	*3.0*	*1.2*	*1.0*
OC-200	Min-Max	2.6–10.4	1.0–1.64	1.0–1.8	1.0–3.4	1.0–2.3	1.0–1.2	1.0–1.1
	Mean	*7.4*	*1.4*	*1.5*	*2.5*	*1.6*	*1.1*	*1.1*
OC-500	Min-Max	1.7–5.4	1.13–1.7	1.0–1.3	1.3–2.4	1.0–1.8	1.1–1.2	1.0–1.2
	Mean	*3.7*	*1.5*	*1.2*	*1.8*	*1.3*	*1.1*	*1.1*
OC-800	Min-Max	1.0–6.6	1.0–1.6	1.0–1.5	1.0–2.2	1.0–1.5	1.0–1.2	1.0–1.1
	Mean	*3.4*	*1.3*	*1.2*	*1.8*	*1.3*	*1.1*	*1.1*
EC-200	Min-Max	6.4–10.9	1.2–1.6	1.4–1.8	2.1–3.3	1.1–2.4	1.1–1.2	1.1–1.2
	Mean	*8.6*	*1.5*	*1.6*	*2.8*	*1.7*	*1.1*	*1.1*
EC-500	Min-Max	2.8–6.8	1.2–1.6	1.2–1.5	1.5–2.66	1.0–1.6	1.0–1.2	1.0–1.2
	Mean	*5.3*	*1.4*	*1.3*	*2.0*	*1.3*	*1.1*	*1.1*
AC-500	Min-Max	1.6–6.5	1.0–1.5	1.0–1.5	1.4–2.7	1.0–1.8	1.0–1.1	1.0–1.3
	Mean	*4.9*	*1.3*	*1.3*	*2.2*	*1.4*	*1.1*	*1.1*

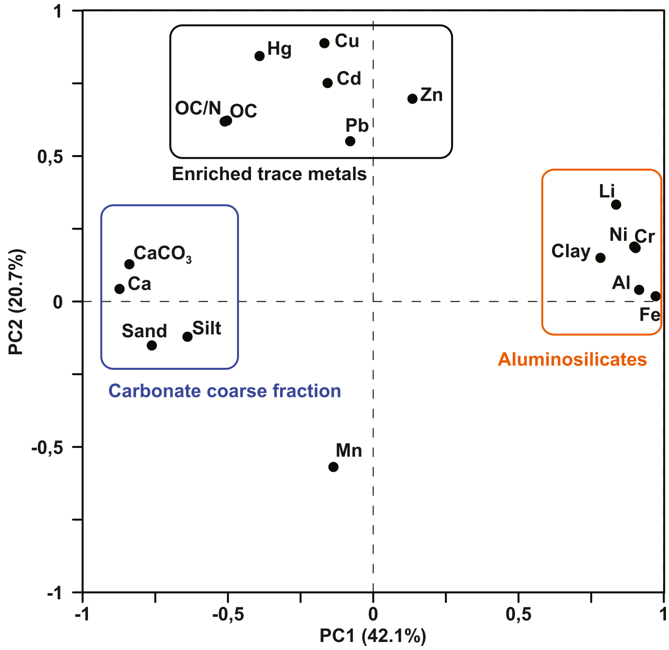

Fig. 5. Principal component analysis performed on the database of the studied variables.

domestic effluents, together with environmental regulations and general decreasing use of some contaminants, have led to a certain reduction of their anthropogenic emissions worldwide (Nriagu, 1996; Pacyna et al., 2001). However, it may take several years or decades until the effects of this reduction can be recognized in the contamination levels of the sedimentary record, mainly depending on sediment accumulation rates and mixing processes (Mil-Homens et al., 2013; Cossa et al., 2014; Palanques et al., 2017).

In the sediments of the continental shelf and the submarine canyons of the Gulf of Palermo, the trend of increasing TM contents was reversed during the 1970s and 1980s (Fig. 4; Table 4). However, there is no evidence of major corrective measures taken in this area during this period that could have generated a change in TM contamination. The causes of the steady decline in environmental quality of the gulf water had not been adequately addressed at the end of the 20th century, (Venezia, 1998) and although the Italian Environmental Ministry included the Gulf of Palermo in the National Remediation Plan in 2002 (Di Leonardo et al., 2007), there is at present still only one waste water treatment plant ("Acqua dei Corsari") that receives half the drainage from the Palermo basin. Therefore, the decrease in TM contamination observed in the sediments accumulated since the 1970s and 1980s cannot be attributed to any protective environmental measures and therefore must have a different cause.

Considering the time in which this change occurred, it may well have been the intensification of engine-powered bottom trawling on the outer shelf and around and inside the canyons that provoked it.

4.3. Effects of trawling on TM contamination of canyon sediments

The scraping and ploughing of the seabed produced by bottom trawling causes increases in near-bottom turbidity due to sediment resuspension (Jones, 1992; Pilskaln et al., 1998; Palanques et al., 2001; Durrieu de Madron et al., 2005; Karageorgis et al., 2005; Dellapenna et al., 2006; O'Neill and Summerbell, 2011) and upward siltation of the underlying seabed sediment because of a preferential redeposition of the coarser silt fraction and advection of the finer silt and clay fractions by currents, following their resuspension (Martín et al., 2014a; Palanques et al., 2014; Paradis et al., 2021b).

In many shelf areas, where natural processes such as storms, riverine sediment discharges and tides can be very energetic, natural sediment dynamics can be more prominent than trawling disturbance, masking its effect on bottom sediments (Bhagirathan et al., 2010; Mengual et al., 2016; Oberle et al., 2016a). On the Mediterranean continental shelves, dynamic processes are usually less energetic than in more open oceanic settings, and sediment resuspension by trawling can be more similar to that induced by natural processes and have a clear impact on the bottom sediment (Palanques et al., 2001; Ferré et al., 2008; Palanques et al., 2014). In deeper slope environments, the trawling effects become even more dominant and persist over larger spatial and temporal scales, as natural sedimentary disturbances are more infrequent (Puig et al., 2012).

The expansion of bottom trawling grounds to the continental slope has occurred at a global scale since the 1960s (Norse et al., 2012; Watson and Morato, 2013; Martín et al., 2014c). In the Mediterranean Sea, trawlers have also increased their engine power, and subsequently, the gear size, the fishing depth and the trawled area per haul (Martín et al., 2008; Sartor et al., 2011), leading to transfer of large amounts of sediment to deeper areas and increasing sedimentation rates on anthropogenic submarine canyons depocentres (Martín et al., 2008; Puig et al., 2012; Martín et al., 2014b and c; Puig et al., 2015; Wilson et al., 2015; Paradis et al., 2018a, 2018b, 2021a).

In the Gulf of Palermo, the expansion of bottom trawling to the slope has also induced the resuspension and transport of large volumes of sediments into the canyons, causing sharp increases in sedimentation rates on their axes and upward siltation since the 1970s and 1980s (Paradis et al., 2021a). All these changes occurred simultaneously with the trend of decreasing TM contamination in the canyon sediments, showing an evident link between the expansion of bottom trawling and the decreasing contamination trend.

The trend of decreasing TM contents since the 1970s and 1980s also coincided with the stronger increasing trends of Ca and $CaCO_3$ and the stronger decreasing trends of clay, Li, Al and Fe (Figs. 2, 3 and 4). This suggests that trawling resuspension also contributed to the increase in coarser carbonate particles, probably with a lower TM content, which settled faster,

Table 4
Decrease in TM enrichment factors (EF) and the percentage decrease in TM concentration (% Dec Conc) from the 1970s-80s.

		Hg	Cu	Zn	Cd	Pb
S-70	$EF_{max} - EF_{surf}$	31.5– 26.1	2.9 – 2.4	2.0 – 1.9	4.8 – 2.4	5.6 – 4.5
	% Dec Conc	*18.9*	*20.0*	*8.2*	*50.5*	*22.4*
EC-200	$EF_{max} - EF_{surf}$	10.9 – 6.7	1.8 – 1.5	1.6 – 1.4	2.4 – 1.2	3.2 – 2.3
	% Dec Conc	*44.4*	*23.9*	*13.5*	*55.4*	*36.7*
OC-200	$EF_{max} - EF_{surf}$	10.4 – 6.3	1.8 – 1.4	1.6 – 1.4	2.3 – 1.1	3.4 – 2.1
	% Dec Conc	*47.6*	*26.0*	*18.7*	*54.6*	*44.5*
EC-500	$EF_{max} - EF_{surf}$	6.5 – 4.3	1.4 – 1.3	1.5 – 1.3	1.6 – 1.2	2.6 – 1.7
	% Dec Conc	40.1	*14.7*	*21.6*	*33.5*	*40.9*
OC-500	$EF_{max} - EF_{surf}$	5.4 – 2.6	1.3 – 1.1	1.7 – 1.5	1.7 – 1.1	5.4 – 2.6
	% Dec Conc	*55.2*	*26.1*	*23.4*	*45.7*	*48.1*
AC-500	$EF_{max} - EF_{surf}$	6.5 – 4.4	1.5 -1.3	1.5 – 1.3	1.8 – 1.3	2.7 – 2.0
	% Dec Conc	*41.4*	*17.9*	*19.2*	*38.3*	*36.7*
OC-800	$EF_{max} - EF_{surf}$	6.6 – 3.2	1.5 – 1.2	1.6 – 1.5	1.5 – 1.3	5.4 – 3.2
	% Dec Conc	*56.5*	*27.2*	*13.8*	*25.0*	*41.9*

and to the decrease in fine-grained aluminosilicate particles with higher affinity to TMs, which were winnowed out.

This demonstrate that bottom trawling plays an important role in reducing the concentrations of TM contamination in canyon axes sedimentary deposits by mixing contaminated with less or non-contaminated sediment and by inducing the winnowing and advection of fine-grained particles and associated contaminants. Trawling could also release TMs into the water column, as observed with other contaminants on trawling grounds (Bradshaw et al., 2012). The decrease in concentrations and EFs of TMs from the maximum in the 1980s until present are shown in Table 4. In general, the decreases were greater in the submarine canyons than on the shelf. The greatest decreases were for Cd on the shelf (50%) and for Hg in the submarine canyons (40%–56%), while the lowest were for Zn, both on the shelf (8%) and also on the canyon axes (13%) (Table 4). The relative decrease in TM concentrations in bottom sediments at 200 m water depth was slightly higher in the Oreto canyon than in the Eleuterio Canyon except for Cd, whereas at 500 m water depth it was clearly higher in the trawled Oreto Canyon axis than in the untrawled Anerella and Eleuterio canyon axes (Fig. 1; Table 4). This suggests that the direct effect of trawling along the Oreto Canyon came in addition to the transfer of resuspended sediment from the surrounding fishing grounds, inducing higher dilution and decreases in TM contamination levels. In addition, the more intense trawling along the Oreto Canyon axis contributed to a further redistribution of resuspended sediment deeper than the operative depth of trawling, to the lower canyon sector (800 m), where the decrease in TM content was still noted.

4.4. Effects of trawling on organic carbon and early diagenesis

A further effect of trawling-induced sediment resuspension is the alteration of OC biogeochemistry (Legge et al., 2020). On fishing grounds, persisting trawling disturbance leads to an impoverishment in OC, given that remineralisation is most efficient when sediments are subjected to alternating cycles of aerobic and anaerobic conditions and that winnowing of the finer particles contributes to the loss of sedimentary OC (Martín et al., 2014a; Oberle et al., 2016b; van de Velde et al., 2018; Paradis et al., 2021b). However, on some continental shelf fishing grounds (Pusceddu et al., 2005; Palanques et al., 2014) and in submarine canyons receiving sediment from adjacent trawling grounds (Martín et al., 2008), an OC increase was also recorded, suggesting that trawling disturbance on the seabed may have different and sometimes opposing impacts that should be taken into account in the sediment carbon budget (Legge et al., 2020).

In the Gulf of Palermo, the uppermost canyon sediments showed variability in OC contents and OC/N ratios (Fig. 2). These changes were more evident in the untrawled Eluterio and Arenella canyons and in the deeper core of the Oreto Canyon (OC-800) and could indicate variable input of natural hemipelagic sediment and sediment resuspended by trawling transferred from the fishing grounds.

The uppermost canyon sediments also showed conspicuous variation in silt and Mn contents and changes in the trends of $CaCO_3$, Al, Fe, Li and Ca (Figs. 2 and 3). In addition, there was a small subsurface Mn peak in the cores taken at 500 and 80 m depth. Similar surface and near-surface Mn peaks were recorded in the Thermaikos Gulf (Karageorgis et al., 2005), in the Var Canyon (Heimbürger et al., 2012) and in the Cap de Creus Canyon (Cossa et al., 2014), but they were not associated with trawling activities in those studies. Mn peaks may be due to formation of autigenic Mn oxyhydroxides at the redox front, where dissolved Mn diffusing upwards from deeper reduced sediment layers meets oxygen diffusing downward from the sediment-water interface. (e.g. Froelich et al., 1979; Haese, 2006; Cossa et al., 2014). The rapid arrival of sediment resuspended by bottom trawling could cause a rapid depletion of O_2 and the dissolved Mn would diffuse upwards, where it would be reoxidized, precipitating as manganese oxide, resulting in high surficial Mn concentrations that diminish rapidly with depth as reducing conditions develop. Further research on the role of trawling on organic matter and redox cycling is needed.

5. Conclusions

The narrow continental shelf of the Gulf of Palermo favours the transfer and deposition of contaminated sediments across the shelf and along the submarine canyons incised in the continental margin. The sediments from the axes of these submarine canyons show an upward-increasing trend of Hg, Pb, Zn, Cu and Cd contamination from the early 20th century, associated with the increase in urbanization and industrial activities.

This trend of TM enrichment reversed to a decreasing trend since the 1970s and 1980s, coinciding with an increase in sedimentation rates and silt, Ca and $CaCO_3$ contents, and also with a strong decrease in clay, Al, Fe and Li contents. These changes occurred simultaneously with the expansion to deeper fishing grounds of bottom trawling fleets equipped with more powerful trawlers (500–2000 hp).

This intensification of bottom trawling has caused the resuspension and transport into the canyons of large volumes of sediments, which were mixed with the canyon sediments, diluting and decreasing their TM contamination levels. Bottom trawling has also generated a preferential deposition of coarser silt and carbonate particles in the canyon axis, whereas the finer terrigenous particles with higher TM affinity were winnowed.

This study provides evidence that sediment resuspension and transport caused by trawling in the Gulf of Palermo have changed TM contamination trends in sediments from the shelf and canyon axes. Trawling dilutes and reduces TM contamination within submarine canyons, but most likely also favours its dispersion and transfer to deeper areas.

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CRediT authorship contribution statement

Albert Palanques: Conceptualization, Funding acquisition, Formal analysis, Visualization, Investigation, Methodology, Resources, Supervision, Writing – original draft, Writing – review & editing. **Sarah Paradis:** Conceptualization, Methodology,Investigation, Data curation, Visualization, Writing – review & editing. **Pere Puig:** Conceptualization, Funding acquisition, Investigation, Methodology, Writing – review & editing. **Pere Masqué:** Conceptualization, Funding acquisition, Investigation, Methodology, Writing – review & editing. **Claudio Lo Iacono:** Conceptualization, Funding acquisition, Investigation, Methodology, Writing – review & editing.

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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