



## Original Articles

# How the reconstruction of faunal communities in a marine protected area (Columbretes Reserve, western Mediterranean) evidence human and natural impacts on fauna

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## ABSTRACT

Reconstruction of marine communities in search of baseline (pristine) conditions is a crucial first step for their future restoration. A recent reconstruction (last century) of the sandy-muddy bottom fauna on the continental shelf of a marine protected area (MPA) was performed in the Columbretes Reserve, including periods after and before the Reserve declaration. The dating of sediments and identification of faunal remains (e.g., shells of benthic bivalves and gastropods and pelagic pteropods) were performed in a core (MC2) at a depth of 87 m in 2018. Radiometric data identified sediments older (below 11 cm) and younger (from the top of the core to 11 cm) than ca. 110 years. Mercury analyses validated the <sup>210</sup>Pb<sub>xs</sub> data at 5–7 cm (1967–1989), with a significant Hg peak that coincided with a period of military activities occurring until 1982 in the Columbretes Islands. Both human and climatic variables affected benthic and pelagic communities. Among the human impacts, the cessation of trawling activity after the declaration of the MPA (1980s) influenced the most dominant benthos (bivalves and gastropods) by i) increases in their abundance and ii) changes in the feeding guilds, with a return to baseline conditions by the increase in filter feeders after trawling cessation vs a high abundance of detritus feeders occurring under high trawling activity. Human activities apparently did not affect diversity levels. In parallel, we also identified some recolonization by Octocorallia since the 1980s. Finally, the increase in the pelagic pteropod *Creses acicula* since 1995 at the MC2 station probably indicates the result of warming of surface waters in recent decades. Our study based on core reconstructions provides for the first time an historical perspective of the impact of trawling on marine benthos and the positive effect of conservation measures in marine protected areas.

## 1. Introduction

Historical reconstructions of marine communities, i.e., chronological studies, have been widely attempted at time scales covering the late Pleistocene and Holocene, sometimes concretely focused on shorter periods (e.g., the last two millennia, Baumgartner et al., 1992; Van de Velde et al., 2019), but there have been few reconstructions focused on recent periods comprising only the last centuries (Valdés et al., 2008; Armenteros et al., 2016; Cartes et al., 2017). Since the greatest human impact on diversity occurred in this recent period, it is

especially important to define a time in this recent past free of such impacts, representing a baseline or 'pristine' reference. Such recent reconstruction of marine communities is crucial as a first step for the future restoration of marine communities. It is necessary to know where a species was naturally distributed before, for example, its reintroduction is being considered. Marine communities have suffered natural fluctuations in recent decades/centuries, both in littoral (Fox et al., 2009; Lirer et al., 2014) and open water systems (Cartes et al., 2017). These fluctuations are responses to natural environmental changes that occur at short-to-medium intervals, e.g., over the last years (warming

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periods in the northeastern Atlantic waters, [Hobson et al., 2008](#); hydrological changes in deep-water masses, [Cartes et al., 2017](#)). In parallel, human impacts (trawling, river damming, delta formation, [Maselli and Trincardi, 2013](#)) have also been identified, appearing in association with natural changes. Distinguishing between the human and natural factors impacting the environment is difficult. Regarding reconstruction studies, the advantage is that some human impacts can be distinguished in time by knowing when they began based on the availability of historical records, and their effects can thus be excluded in the oldest periods. In marine systems, human influences on communities have been obvious, especially in littoral/shelf areas, which have been exploited and modified over centuries. The effects of trawling on seabed communities are a major issue worldwide ([Pitcher et al., 2022](#)). Deep sea areas, however, have been globally less impacted and more recently impacted than shelf areas. In the case of the deep Mediterranean, in which deep sea trawling (at > 400 m) has focused on aristeid shrimp, trawling began only from the 1940–50 s ([Oliver, 1953](#)), even starting later in some specific areas ([Cartes et al., 2013, 2017](#)).

Reconstructions have been performed in a variety of terrestrial and also marine environments, in this case ranging from coastal lagoons ([Van de Velde et al., 2019](#)) and estuaries/bays ([Armenteros et al., 2016; Vidovic et al., 2016](#)) to, more scarcely, open sea systems. Reconstructions in the open sea have been performed on pelagic fish such as anchovies ([Baumgartner et al., 1992; Valdés et al., 2008](#)) and on epimesopelagic fish ([Salvatteci et al., 2019](#)) by recording fluctuations in scales and other fish remains, such as vertebrae, bones ([Valdés et al., 2008; Salvatteci et al., 2019](#)) and otoliths ([Cartes et al., 2017, 2018; Agladi and Albano, 2020](#)), and in the case of sharks using dermal denticles ([Dillon et al., 2017](#)) buried in sediments. Otoliths are more resistant and have better species-specific morphology than scales or bones. It is possible, based on otoliths, not only to determine the fish species ([Girone et al., 2010](#)) but also its size ([Cartes et al., 2018](#)), thus reconstructing ecological changes in teleost fish assemblages. Reconstructions have also been conducted based on benthic shelf invertebrates, e.g., on benthic molluscs ([Armenteros et al., 2016; Gallzemer et al., 2017](#)). Parallel dating of sediment cores by radionuclide analyses, as performed in [Valdés et al. \(2008\)](#), [Armenteros et al. \(2016\)](#) and [Cartes et al. \(2017\)](#), can be used to identify the timing of biomass/diversity shifts, which can then be related to recent climatic events or human impacts. Even so, sedimentary environments can often be affected by bioturbation and postdepositional geophysical processes.

Shifts in species community composition can be related to indices of climatic variability (e.g., the North Atlantic Oscillation, NAO) or climatic events such as ENSO (El Niño-Southern Oscillation). Obviously, such relationships can only be attempted for periods in which information on such climatic indices/variables is available, i.e., covering the last few hundred years at most. Shifts are also related to fishery impacts, which especially affect sessile species, such as bamboo corals ([Cartes et al., 2017](#)), and to pollution ([Armenteros et al., 2016](#)). Since data on the main human impacts, such as trawling efforts or river damming (through changes in river discharges), are also basically available starting from the beginning of the last century, recent paleobiological analyses, such as those presented here, are only possible and have focused on the last centuries (basically SXX, SXXI). These kinds of studies are very scarce in recent times, and even more so in marine protected areas (MPAs). In MPAs, we know with precision in what year human activities were interrupted, so we can expect some recovery of fauna and in its environment.

For dating marine sediments in this recent period, the most widely employed method is to establish  $^{210}\text{Pb}$  ( $^{210}\text{Pb}_{\text{xs}}$ ) profiles in sediment cores ([Appleby and Oldfield, 1978; Koide et al., 1972; Robbins and Edgington, 1975; Sánchez-Cabeza et al., 2012](#)), while studies focused on older periods (the scale of millennia) can combine different dating methods, such as radiocarbon dating ([Tomašových et al. 2017; Van de Velde et al., 2019](#)). The  $^{210}\text{Pb}$  isotope is a natural radionuclide with a half-life of approximately 22 years and is a byproduct of  $^{226}\text{Ra}$ , which is

scarce in the ocean.  $^{210}\text{Pb}$  is supplied to the aquatic environment by atmospheric deposition and is rapidly scavenged by particles to the seafloor; thus,  $^{210}\text{Pb}_{\text{xs}}$  is ideal as a tracer for dating aquatic sediments deposited in the last ~ 100 years. Since  $^{210}\text{Pb}$  dating requires some a priori assumptions, the depth-age correspondence needs some independent time marker to be supported.  $^{137}\text{Cs}$  is a commonly measured, man-made gamma-emitter globally present in the environment since nuclear bomb testing in the atmosphere in the 1950 s and 1960 s ([UNSCEAR, 2008](#)). However, other techniques can provide clues on distinct periods occurring during the recent history of an ecosystem, as is the case for heavy metals or metalloids (i.e., Hg and Pb), which can indicate, for example, when industrial and other anthropogenic activities began ([Armenteros et al., 2016; Sánchez-Cabeza et al., 1993](#)) or even when their concentrations changed from one type to another type of anthropogenic source ([Prego et al., 2008](#)).

The reconstruction of systems that have been altered by human impacts is a first step in the restoration of such systems, allowing the recovery of the previous state of biodiversity. Knowledge about the structures of pristine communities is of great importance for the establishment of reference levels and thresholds in studies evaluating the environmental status of benthic habitats using indicators, such as the studies carried out in the Marine Strategy Framework Directive, MSFD (2008/56/EC), and the Habitats Directive, enhancing measures to protect and preserve coastal and open ocean ecosystems through adequate policies.

In recent decades, shelf areas have benefited from some forms of protection, especially with the increasing designation of marine protected areas since the 1960 s-70 s ([Humphrey and Clark, 2020](#)) and the implementation of the MSFD (2008/56/EC). The Columbretes Islands Marine Reserve, located on the Mediterranean coast of Spain, is a good example. This archipelago has a set of four groups of volcanic islands located to the east of the Iberian Peninsula. A law passed in December 1987 regulated the management to protect the islands; the area was declared a Natural Park in 1988 and a marine reserve (with 4400 ha, one of the largest in Spain) in 1990, with other considerations such as being a Special Protection Area for Birds (SPA) under the Habitats Directive (92/43/EEC). As a result, trawling was forbidden, and fisheries were reduced to some traditional nondestructive activities.

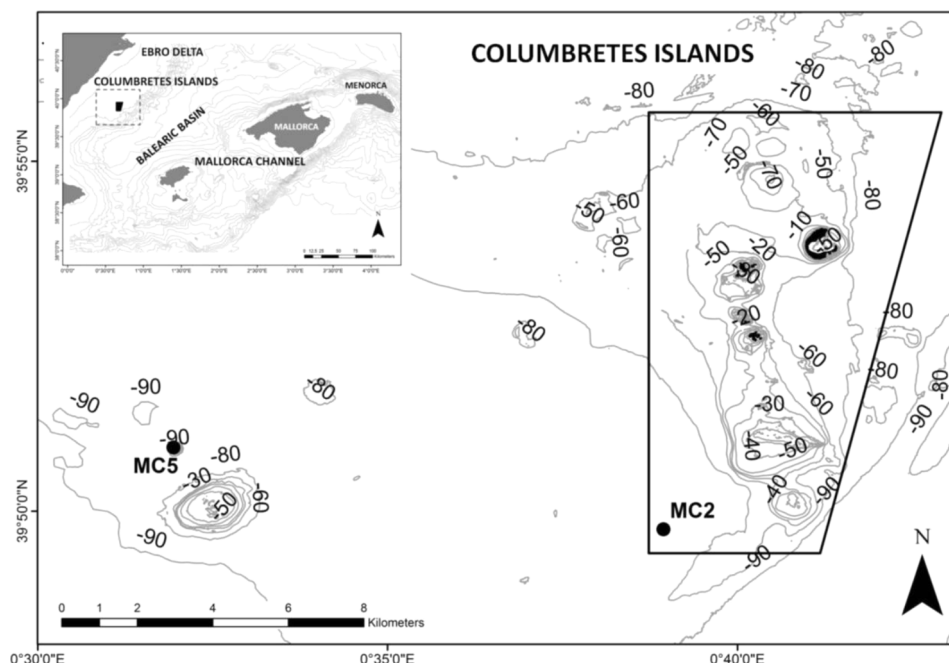
The aim of our study was to analyse recent natural and human-induced changes, including fishery exploitation, in the benthic and benthopelagic communities dwelling on the continental shelf (87 m depth) of a marine protected area, the Columbretes Archipelago, based on the reconstruction of recent assemblages in a sediment core (CV-MC2). We analysed i) recent temporal shifts in the composition of the thanatocoenosis and ii) the possible influence of long-term environmental variables on different faunal compartments (zooplankton, benthos).

## 2. Materials and methods

### 2.1. Sampling

The Columbretes Archipelago is located on the continental shelf off the eastern coast of the Iberian Peninsula (Balearic Basin, [Fig. 1](#)). This archipelago is composed of four groups of volcanic islands located 56 km to the east of the mainland coast (Cape of Oropesa, Iberian Peninsula). The study area is in the Marine Reserve of Columbretes (39°49' to 39°55' N, 0°40' to 0°42' E), with a surface area of 5543 ha; it was declared a protected area in 1987, a Natural Park in 1988, and a marine reserve in 1990. Since then, trawling has been forbidden at the station where multicorer 2 (CV-MC2) was used to collect sediment cores in the southern area of the archipelago, SW of Bergantín (*El Carallot*) Island.

During cruise COLVIDEO0518 (21–27 May 2018, project MAR-EMATING), a sediment core (CV-MC2-T1) was collected at a depth of 87 m at 0° 6496 E, 39° 8290N over a sandy-muddy sea bottom with biogenic remains (bivalves, gastropod shells). The area, situated on the



**Fig. 1.** The study area (Balearic Basin on the square map) over the continental shelf of the Levantine coasts of the Iberian Peninsula, indicating stations sampled (●), and the specific MC2 core (87 m depth) into the Reserve (framed area) that was analysed. The location of MC5, at the neighbouring shelf to MC2 also indicated.

continental shelf, on one of the widest shelves in the western Mediterranean, is flat, and no slopes or canyons with steep walls are located close to station MC2 (Fig. 1). MC2 was collected within the marine reserve, and historical reconstruction is the focus of the present study. The area surrounding station MC2 is homogeneous enough so that the MC2 results can be extrapolated to a wider area at similar depths, at least within the reserve (see discussion). The CV-MC2 was taken on board the R/V *García del Cid*.

We used a 6-tube (tube  $\varnothing = 10$  cm) multicorer. Sediments (and macrofauna, both dead remains and alive) were sampled. One core (tube) of MC2 (MC2-T1) was analysed and cut every 1 cm to a depth of 16 cm for radionuclide, mercury, microplastics and faunal analyses. Since the sediment consisted of sandy mud with biogenic remains, it was not possible to further drive the core into the sediment to obtain samples deeper than 16 cm. The sediment slices were frozen at  $-20$  °C onboard the vessel in individual, sealed plastic bags until the laboratory analyses were conducted. From each MC2 slice, approximately 20 g of wet weight was separated for the radiometric analysis, approximately 1 g was separated for Hg analysis, and the rest (between 83.2 and 128.0 g) was used for microplastic and faunal analyses.

The reconstruction of dead fauna in a sediment core generates a sort of temporal series of the thanatocoenosis identified. Aspects such as to what extent the results obtained can be extrapolated and sampling replicability were considered/assumed as follows: i) the information integrated in each 1-cm level (representing a number of years) is independent as the observation of each other level (slice) analysed; ii) the analysis of a single core is sufficient in such reconstruction studies; without replicability between parallel tubes of the multicore (see, e.g., Valdés et al., 2008; Margaritelli et al., 2016; Vidovic et al., 2016; Cartes et al., 2017). The tubes (replicates) for dead fauna (separated by ca. 1 m) may have a lower spatial variability than the variability (patchy distribution) observed for living benthos, since living animal moves for example to feed, for reproductive migrations or settlement. In this way, a preanalysis performed in four tubes of MC2 evidenced a high similarity (range: 76–80.8 % between the dead fauna of benthic molluscs, Bray Curtis index) at the most superficial level (0–1 cm) of the core. We should expect that the 0–1 cm level must be subjected to a higher variability (so, less similarity) than the levels distributed deeper in a

(organized) core. These considerations further confirmed the approach adopted.

## 2.2. Dating (radionuclid) analysis

Radionuclide measurements were performed at ENEA Research Centre, Sta. Teresa (Italy) by gamma spectrometry using ORTEC High Purity Germanium detectors with the following performance specifications based on the standard geometry. The resolution and relative efficiency of the detectors used were 1.90 keV and 17 % for Detector 1 and 2.30 keV and 60 % for Detector 2. The resolution for the 662-keV  $^{137}\text{Cs}$  gamma emission was 1.8 keV. Each 1-cm sediment sample from MC2 (16 cm long) was wet-weighted and then dried at 50 °C to a constant weight to calculate the porosity and mass vs depth. The dried samples were ground in an agate mortar, and containers with standard geometry were filled with approximately 20 g of sediment. Then, the sediment was stored for at least 22 days to ensure that the short-lived daughter product,  $^{222}\text{Rn}$ , had decayed to secular equilibrium with  $^{226}\text{Ra}$ . The  $^{210}\text{Pb}$  measurements were corrected for self-absorption (Cutshall et al. 1983). The minimum detectable activities (MDA) were estimated. Calibration for  $^{137}\text{Cs}$  was performed using the Eckert & Ziegler Analytics reference standard QCYA48. Quality controls were routinely made by analysing the IAEA-385 reference material. Detector blanks were determined from measurements performed on empty sample containers over a few days (Carroll and Lerche, 2003; Zaborska et al., 2007).

To fit the obtained  $^{210}\text{Pb}$  profile, a CRS model was adopted.

## 2.3. Dating validation: Hg and microplastic analyses

The sediments were dried in the laboratory at room temperature (approximately 20–25 °C) to a constant weight, ground using plastic mortars and sieved through a 63- $\mu\text{m}$  plastic mesh. The total mercury concentrations of the samples were directly determined using an Advanced Mercury Analyser (LECO AMA-254), with a detection limit of 0.01 ng Hg. The sediments were dried at 120 °C for 70 s and thermally decomposed between 550 and 750 °C for 200 s with an oxygen carrier flow. The decomposition products were carried by an oxygen flow (99.99 % purity) to an Au amalgamator. Selectively trapped mercury

was released from the amalgamator by a brief heat-up at 850 °C and finally quantified (measuring cycle of 45 s) as HgO by cold vapour AAS at 253.65 nm (Method 7473, US EPA Methods). The sediments were measured in triplicate, and quality control was assessed by analysing blanks and Certified Reference Materials (MESS-3, marine sediment reference material), obtaining recoveries of approx. 99 %.

The microplastics (balls of fibres) present in each level of sediment were sorted under a stereomicroscope (x10-x40) and counted in each 1-cm layer. Since it was impossible to treat the sediment (e.g., cutting cores onboard the vessel or filtering sediments onboard for faunal analyses) while avoiding airborne contamination, we did not consider the occurrence of isolated small and light fibres that could be introduced to the sediments through the air (Carreras-Colom et al., 2020).

#### 2.4. Analysis of the thanatocoenosis (biological remains)

The remaining sediment from each 1-cm sediment slice/level (between 83 and 128 g) was sieved through a 200- $\mu$ m sieve to isolate the remains of the fauna in MC2, with the exception of benthic molluscs, which were sieved gently through a 500- $\mu$ m sieve. This allows the removal of fine particles from solid biological remains. Each level was analysed randomly (without following any predetermined order) under a stereomicroscope (x10-x40), and remains were sorted with forceps and identified to the lowest possible taxonomic level. We distinguished the following items: i) Living benthic meiofauna, which was classified to the lowest taxonomic level; ii) Biological remains of bivalves and gastropods, classifying the shells whenever possible (see below); iii) Fish otoliths and other fish remains (scales, vertebrae). The otoliths were identified and measured with the help of an ocular micrometer attached to a stereomicroscope. The otoliths could be counted, while the other fish remains (scales, vertebrae), which were less frequently found, were assigned to a maximum of one individual; iv) The shells of pteropods (up to 6 species identified belonging to pelagic fauna: zooplankton) that were counted; v) The remains (sclerites) of gorgonians, also identified and quantified, since coral sclerites rarely appeared broken; and vi) The chelae of crabs and other calcified decapods that were identified and counted. It was possible to know the number of specimens (crabs) in the sample when the chelae were clearly of different sizes and were hence attributable to different individuals. Bivalves and gastropods were identified and quantified after being passed through a 500- $\mu$ m sieve. The specimens were quantified as representing one individual when the shell was > two-thirds intact and included the apex and spires of gastropods and umbo/hinges in bivalves. This taphonomic criterion related to fragmentation is within/above the standards applied in paleobiological studies (see details in Table S1). We did not count the broken shells that had higher probabilities of being reworked, as they likely did not have the same estimated age of the sediment where they were immersed. The most abundant species (or those indicating tendencies in CCA analyses) were assigned to basic trophic guilds and habitat preferences based on bibliographic (Dame, 1996; Rueda et al., 2009; Armenteros et al., 2016) or other sources on molluscan feeding habits (Todd, J.A., <https://fossils.its.uiowa.edu/database/mollusc/mollusclifestyles.htm>). The classification of species in feeding guilds requires simplification, as species are capable of following more than one feeding strategy. Thus, members of the Subclass Paleotaxodonta, the most primitive living bivalves, have short siphons and are mainly considered surface deposit feeders (detritivores). Although filter feeders can show different levels of filtering efficiency (e.g., *Corbula gibba* can be twice as efficient as *Cerastoderma edule*, or *Aequipecten* spp., Kjørboe and Møhlenberg, 1981; Dame, 1996), all were simply considered filter feeders in our approach. Gastropods may also exhibit a variety of feeding habits, including suspension feeding, scavenging, detritivory, and carnivory (Taylor, 1998). Small Iravadiidae (*Hyala* sp.) are subsurface deposit feeders (<https://en.linkfang.org/wiki/Hyala>), as are Rissoidae.

#### 2.5. Treatment of the biological data

##### 2.5.1. Changes in abundance with core depth

We counted the number of living organisms and dead remains of fauna (e.g., bivalves, benthic gastropods, fish otoliths, sclerites, and pteropods) at each 1-cm level, standardized to the sediment wet weight. Smaller taxa (e.g., foraminiferans) were not considered. The data matrix generated had 113 taxa/categories.

In order to identify possible changes in the dynamics and structure of the dead communities analysed the abundance and the diversity of more abundant/representative taxa were calculated per each core level. Standardized abundances (n°/100 g sediment) were plotted by depth in MC2 within the core series for the selected dead (subfossil) taxa: i) pteropods of the main 3 species *Clio pyramidata*, *Cavolinia inflexa*, and *Creseis acicula*; ii) sclerites of corals (mainly *Octocorallia*, including soft corals, *Alcyonacea* and sea pens, *Pennatulacea*) and iii) the most abundant species of bivalves (*Parvicardium minimum*, 28 % of total bivalves; *Timoclea ovata*, 23 %; *Saccella commutata*, 10 %; *Hiatella arctica*, 6 %), and gastropods (*Turritella communis* 23 %; *Calyptrea chinensis* 14 %). Shannon diversity ( $H'$ , Magurran, 2013) was also calculated and plotted by depth for i) the whole benthic community, mainly composed of bivalves, gastropods, fish and decapods (all representing 81 % of the benthic community); and ii) fish (*epi-meso* and benthic species) and decapods, representing predators/scavengers. 1-way ANOVA and post-hoc tests were applied to check changes in the abundance of some dominant species. The 3 levels compared (benthos) were: 0–5 cm, the period of Reserve declaration; 6–9 cm, the period with the highest fishing activity, and older (>10 cm) layers, periods without fishing activity.

##### 2.5.2. Changes in the composition of the thanatocoenosis

Changes in the composition of the thanatocoenosis (abundance in each 1-cm sediment level) were analysed by multidimensional scaling (MDS) analysis, a multivariate ordination technique. MDS analysis was performed using the Bray–Curtis index (after log-transformation of the data). Taxa/species with low frequencies of occurrence (one or two occurrences) were removed from the matrix. The analysis was performed for the main compartments zooplankton and benthos, which is expected show a different response to environmental changes. We analysed: i) the *epi-mesopelagic* assemblages (pteropods and few epipelagic and mesopelagic fish); and ii) the whole benthic community (as described above). Sclerites of corals (*Octocorallia*), together with other remains (e.g., echinoid carapace pieces, sponge spicules, very fragile and often broken), could not be attributable to a concrete number of individuals, so they were not considered in MDS data matrices.

PERMANOVA tests (distance-based permutational multivariate analysis of variance; Anderson et al., 2008) were performed on the same abundance matrices (for zooplankton and benthos) in each 1-cm MC2 core. The PERMANOVA design was based on the factor "depth" in the MC2 core, with 4 groups *a priori* established (A, B – with B1, B2 - and C). Group A comprised the 0–1-, 1–2-, 2–3- and 3–4-cm levels. Group B included the levels 4–5-cm, 5–6-cm, and so on to 11 cm, the lowest level in which some Pb<sup>210</sup> signal was detectable. Finally, Group C included the rest of the levels to the deepest level sampled in MC2 (16 cm), where the Pb<sup>210</sup> signal disappeared in the radionuclide analyses. Group B was subdivided into 2 subgroups, B1-B2, each with a similar number of levels (B1: 4–5-, 5–6-, 6–7-, and 7–8-cm; B2: 8–9-, 9–10-, and 10–11-cm).

##### 2.5.3. Relationship with environmental variables

Finally, the relationships among (changes in) the density of the main community components and possible explanatory environmental variables (objective 2) were explored by multivariate canonical correspondence analysis (CCA: Ter Braak, 1986). CCA is a multivariate method for which environmental relationships/gradients can be identified from ecological data (Ter Braak and Verdonschot, 1995) by the joint distribution (plotting) of species and environmental variables on ordination



axes. The data fit well to CCA conditions (unimodality and wide gradient for ordination axes). Data were log-transformed and XLStat software (AddinSoft Inc.) was used for these analyses. Different CCAs were performed based on the zooplankton and benthos abundance matrices. Again, species belonging to zooplankton or benthos have different life strategies and trophic roles (e.g. trophic level, origin of food consumed), to analyse them by separate gives a different view of the community dynamics/response. An initial CCA was performed on the zooplankton groups (pteropods and mesopelagic fish), and the rest of the CCAs were conducted on bivalves, gastropods and nonmollusc (mainly fish and decapods) benthic species, hence considering taxa situated at different levels of the food web, from filter feeders to top predator/ scavengers. The CCA considered the chronology of the sediment (as periods or intervals of years), as shown in Table 1 (see also the radionuclide results), as well as the possible explanatory variables attributable to each period associated with each 1-cm layer over approximately the last hundred years. Analyses considered those levels assigned to an age by radionuclide dating, which comprised 10 levels since the year 1927. Such variables included i) the overall temperature in the water column (mean T), from the surface to a depth of 50 m (0–50 m), corresponding to surface waters (SW) and to the nearest MC2 station level (100 m); ii) the same for salinity ( $\text{g kg}^{-1}$ ), measured both throughout the water column and near the sea bottom; iii) the NAO climatic index (North Atlantic Oscillation), as the mean of the NAO indices for all of the periods established for a given core layer; the winter NAO indices were also considered; iv) Ebro River runoff data (mean of annual discharges in the study period), as the plume effect caused by river runoff that may arrive to the shelf in which the Columbretes Islands are located; and v) fishery efforts, based on the elementary variable (mean) number of boats in the nearest port to the MC2 area (Castellón). These variables were selected, in addition to their environmental and climatic interest, due to their availability (e.g., T and S data in the water column available since 1900). The explanatory variables were preanalysed by calculating the Pearson correlation coefficients among all variables. The existence of intercorrelated variables is not an obstacle to perform a CCA, but can be less clear for the interpretation of results. In our case, we prefer to avoid some redundant variables showing a significant relationship ( $T_{100\text{ m}}$  and  $S_{100\text{ m}}$ ,  $r = -0.93$ ; NAO and winter NAO,  $r = 0.88$ ). We also found a significant inverse correlation between the Ebro discharge (runoff) and winter NAO ( $r = -0.68$ , Spearman rank  $\rho$ ). All variables were expressed as the means of the annual values in the period established for each core slice. Temperature (T) and salinity (S) data were downloaded from the MEDATLAS database for the area of the Balearic Basin (M. Vargas-Yañez, I.E.O. Málaga). Ebro flow data ( $\text{m}^3/\text{s}$ ) were obtained from the *Confederación Hidrográfica del Ebro* (<https://www.chebro.html>), and the annual North Atlantic Oscillation (NAO) indices were downloaded from the Climatic Research Unit of East Anglia University (Norwich, UK: <https://www.cru.uea.ac.uk>).

**Table 1**

Geochronology of the MC2 sediment core at 87 m based on radiometric analyses and the Constant Rate of Supply (CRS) dating model. Years of each sediment layer are indicated. According to CRS model, MAR is calculated at the bottom section of each layer.  $u(\text{MAR})$  is uncertainty.

Layer (cm)	Years (a)	MAR * ( $\text{g cm}^{-2} \text{ a}^{-1}$ )	$u(\text{MAR})$ ( $\text{g cm}^{-2} \text{ a}^{-1}$ )
0–1	2013–2018	0.21	$\pm 0.03$
1–2	2007–2013	0.18	$\pm 0.03$
2–3	2002–2007	0.19	$\pm 0.04$
3–4	1995–2002	0.14	$\pm 0.03$
4–5	1989–1995	0.17	$\pm 0.04$
5–6	1980–1989	0.12	$\pm 0.02$
6–7	1967–1980	0.09	$\pm 0.02$
7–8	1954–1967	0.08	$\pm 0.01$
8–9	1927–1954	0.04	$\pm 0.01$
9–10	1879–1927	0.02	$\pm 0.01$
10–11	<1908		

### 3. Results

#### 3.1. Radionuclide ( $^{210}\text{Pb}$ ) sediment dating

Numerical specific activities can be found in Table S2. Fig. 2 represents the downcore activity profiles of  $^{210}\text{Pb}_{\text{xs}}$ . The profile of  $^{210}\text{Pb}_{\text{xs}}$  shows a decreasing trend with core depth, although this trend does not follow an exponential decay. Therefore, in the MC2 sediment core, the constant rate of supply (CRS) dating model was applied (Appleby and Oldfield, 1978; Carroll and Lerche, 2003; ) instead of CF:CS.

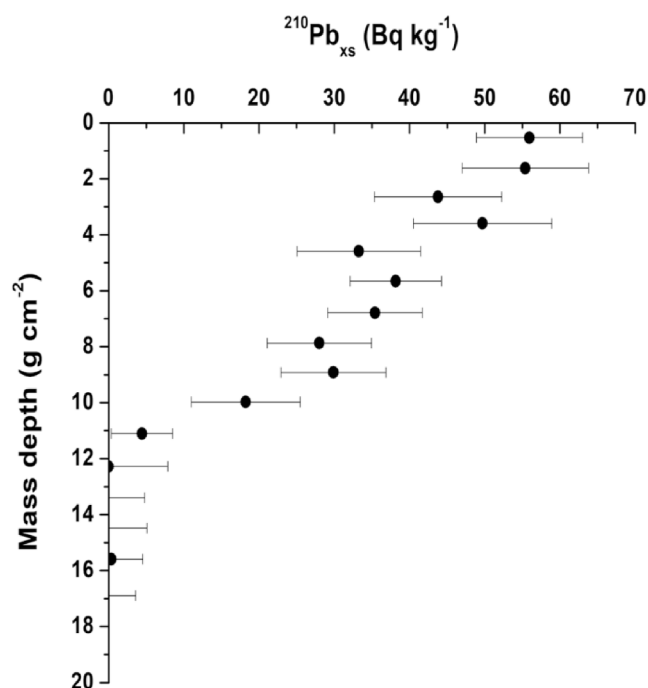
The age (period between sediment deposition and sampling) uncertainty calculated with the CRS model in the MC2 core is 10 %. Table 1 presents the years to which each sediment layer in the MC2 sediment core was attributed with the calculated MAR values and their uncertainties.

Although to corroborate the  $^{210}\text{Pb}_{\text{xs}}$  chronology,  $^{137}\text{Cs}$  activity was also measured in the core, the activities were too close to MDA to properly identify a time marker (Drexler et al., 2018).

#### 3.2. Dating validation results

##### 3.2.1. Hg results

The mercury concentrations oscillated between 9.4 and 23.7  $\mu\text{g}/\text{kg}$  along the sediment core (Table S3, Fig. 3). Constant levels ranging from 9.4 to 12.1  $\mu\text{g}/\text{kg}$  were found from depths of 15 to 11 cm, with a continuous increase from 11 cm (9.8  $\mu\text{g}/\text{kg}$ ) to 6 cm (23.7  $\mu\text{g}/\text{kg}$ ). This period of increase corresponds to the years between approximately 1910 (110 years ago, by  $^{210}\text{Pb}$  extinction) and 1980. At depths of 5–6/6–7 cm, Hg reached a maximum (23.7  $\mu\text{g}/\text{kg}$ ), corresponding to the periods 1967–1980 and 1980–1989, respectively. Once this maximum was reached, Hg showed an almost stable concentration (21.2–23.6  $\mu\text{g}/\text{kg}$ ) between depths of 6 and 2 cm (years 1980 to 2007), decreasing in the upper layer below 20  $\mu\text{g}/\text{kg}$  (years 2013–2018), with a peak at a depth of 1–2 cm (2007–2012). So, the Hg peaks at 6–7 and 5–6 cm, corresponding to the periods of 1967–1980 and 1980–1989 and coinciding with the period of military activities in Columbretes, validated  $^{210}\text{Pb}_{\text{xs}}$



**Fig. 2.** The  $^{210}\text{Pb}_{\text{xs}}$  activity profile along the mass depth scale of the MC2 sediment core (collected at a depth of 87 m off the Columbretes Islands). Error bars represent 1-sigma uncertainties. Layers deeper than 10 cm are older than 1908.

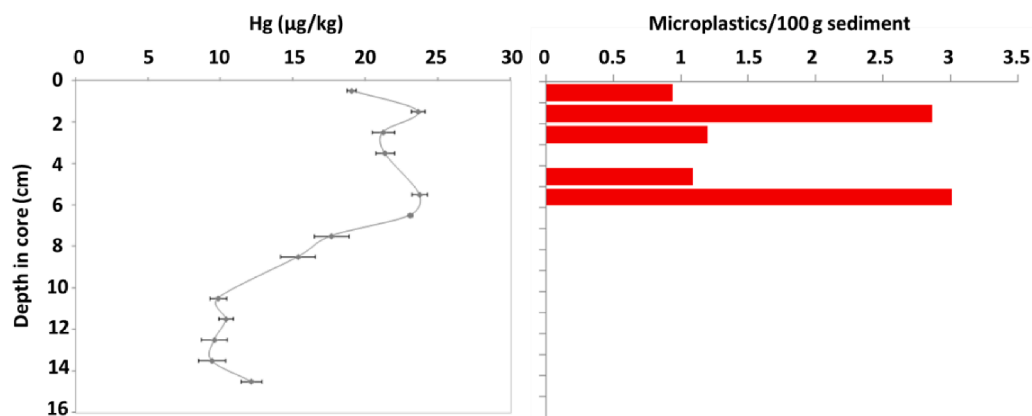


Fig. 3. Mean concentration in the MC2 core related to core depth (cm) of: i) mercury, Hg (µg/kg) with standard deviation (left) and ii) microplastics (fibre balls; isolated fibres excluded) per 100 g of sediments.

dating, as did also the Hg increase at the beginning (before the first half) of SXX, matching the beginning of industrialization in the study area.

3.2.2. Microplastic results

Microplastics (balls of fibres) were found up to the 6- to 7-cm layer (6.79 g cm<sup>2</sup>), corresponding to the year 1967 +/- 4 years (Fig. 3). We considered the occurrence of microplastics (large balls of fibres, not isolated fibres that can occur by airborne contamination) to fit the <sup>210</sup>Pb<sub>xs</sub> chronology. The first findings of microplastics in MC2 sediment core validated also the <sup>210</sup>Pb<sub>xs</sub> dating results (see Discussion) as already confirmed by the Hg profile.

The chronology of all these markers, associated with concrete events, are resumed in Fig. 4,

3.3. Analysis of fauna

3.3.1. Living macro-meiofauna (benthos)

The living fauna identified in MC2 was rather scarce. Only seven different living taxa were found in the first few sediment sections, with 26 specimens counted. Practically all living organisms (>90 %) found were collected at depths of 0–4 cm in MC2; these organisms were benthic and of small size (rather meiobenthos), so considering their low abundance (nematod abundance was only 1.5 ind/10 cm<sup>2</sup>) possible contribution to the bioturbation of the upper part of core sediments would be low. >45 % of the living fauna were nematodes (part of meiofauna), small polychaetes represented 29.4 % of specimens, while small crustaceans such as the amphipod *Monoculodes* sp., tanaids, the cumacean *Diastylodes bacescoi* and Harpacticoida were represented by single individuals (Table 2).

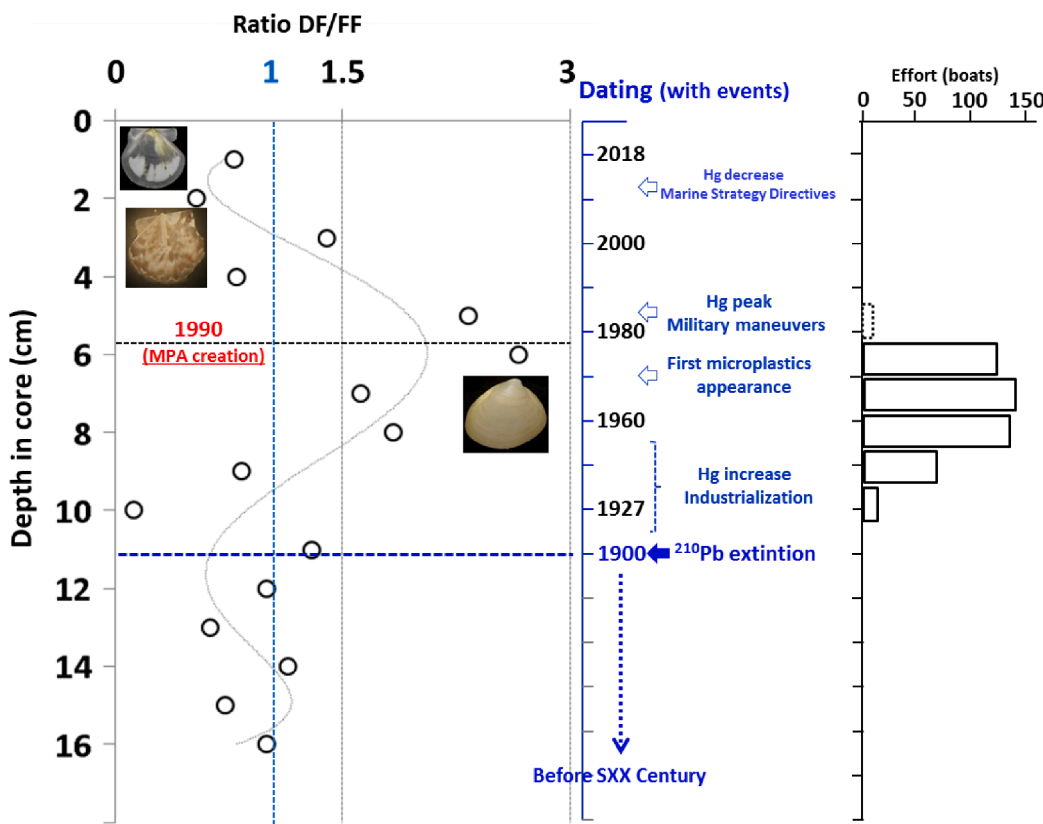


Fig. 4. Chronology for dating of the MC2 sediments, including the (approximate) date of the markers (Hg concentration and first microplastic occurrence) associated to some concrete events, used to validate the <sup>210</sup>Pb analyses. The estimated effort (n° boats in the area) also included. The scheme of the evolution of benthic bivalves as a function of the trophic guild adopted in MC2 station was also included considering those species occupying the extreme positions in the CCA (Fig. 10c): i) Deposit feeders DF (*Nucula nitidosa*, *Nucula* sp.; *Bathycarca* sp.); ii) filter feeders, FF (*Palliolium* sp.). Vertical line is representing a theoretical ratio between DF and FF. Reserve declaration year (1987–1990) is indicated by the upper discontinuous line.

**Table 2**  
Living fauna and faunal remains (number per core level/100 g sediment) of different taxa found in the sediment columns of MC2 off the Columbretes Islands. L, sediment core level; WW, weight per sample. Trophic group (TG) for bivalves/gastropods in the CCA: F, filter-feeders; D: deposit feeders; C: carnivores; S: with symbiont bacteria.

CV0518-MC2-T1	L	0-1 cm	1-2 cm	2-3 cm	3-4 cm	4-5 cm	5-6 cm	6-7 cm	7-8 cm	8-9 cm	9-10 cm	10-11 cm	11-12 cm	12-13 cm	13-14 cm	14-15 cm	15-16 cm
87,3 m	WW	106.35	104.65	83.24	90.30	91.96	99.64	102.91	88.16	94.62	88.97	102.26	99.98	107.64	88.42	101.43	128.01
<b>Alive fauna</b>																	
Nematoda		6	2	2	1	0	1	0	0	0	0	0	0	0	0	0	0
Polychaeta (unid.)		3	1	3	0	0	0	0	0	0	0	0	0	0	0	0	0
Harpacticoida		0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0
<i>Monoculodes</i> sp.		1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
<i>Diastylodes bacescoi</i> Fage, 1940		1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Tanaididae		3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Cypridinidae		1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
<b>Dead Fauna/Remains</b>																	
<b>Polychaeta</b>																	
<i>Glycera</i> spp.		1	1	0	1	1	1	2	1	1	1	1	0	2	3	1	2
Nereidae		0	0	0	1	0	0	0	0	0	0	0	0	0	0	1	0
Polychaeta remains (setae...)		0	5	0	1	3	0	0	0	0	0	1	0	0	0	0	0
<b>Crustacea</b>																	
<i>Alpheus glaber</i> (Olivi, 1792)		1	0	1	0	0	2	0	0	0	0	0	0	1	2	0	0
Palinuridae		0	0	0	0	0	1	0	0	1	0	0	0	0	0	0	2
<i>Ebalia</i> spp.		5	5	6	4	5	3	9	3	5	3	7	3	4	6	17.7	9
Brachyura Portunidae		0	0	1	1	0	0	2	0	0	1	0	0	3	1	1	1
Brachyura ( <i>Monodaeus couchi</i> , Couch 1851)		1	1	0	0	2	1	0	0	0	0	0	0	0	0	3	1
Brachyura (unid.)		1	0	0	0	0	0	0	1	1	1	0	1	0	0	0	0
<i>Apseudes spinosus</i> (M.Sars, 1958)		0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0
<i>Scalpellum scalpellum</i> (Linnaeus, 1767)		0	0	1	0	0	0	0	0	0	0	0	0	1	0	0	0
<b>Mollusca (Bivalvia)</b>																	
<b>TG</b>																	
<i>Nucula hanleyi</i> Winckworth, 1931	D	1	0	0	1	2	0	0	0	0	0	1	0	1	1	2	0
<i>Nucula nitidosa</i> Winckworth, 1930	D	0	1	0	0	1	1	2	1	1	0	1	1	0	0	0	3
<i>Saccula commutata</i> (Philippi, 1844)	D	18	23	19	25	21	18	15	23	23	16	20	21	26	23	40	41
<i>Arca tetragona</i> Poli, 1795	F	1	0	0	1	2	0	3	1	1	0	0	0	0	2	0	3
<i>Batharca pectunculoides</i> (Scacchi, 1835)	D/F	10	6	7	7	18	7	8	9	8	0	12	8	5	6	16	14
<i>Anadara gibbosa</i> (Reeve, 1844)		2	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0
<i>Rhomboidella prideauxi</i> (Leach, 1815)		0	1	1	0	0	0	1	0	1	0	0	0	0	0	1	1
<i>Modiolula phaseolina</i> (Philippi, 1844)	F	0	3	3	2	2	2	2	0	1	1	2	2	2	0	2	5
<i>Musculus subpictus</i> (Cantraine, 1835)	F	0	3	0	0	0	3	1	4	1	0	3	0	2	0	1	3
<i>Similipecten similis</i> (Laskey, 1811)	F	14	11	5	7	8	3	6	6	12	6	9	8	6	7	22	16
<i>Aequipecten opercularis</i> (Linnaeus, 1758)	F	2	2	4	1	5	0	2	3	3	1	5	2	1	2	0	6
<i>Palliolium incomparabile</i> (Risso, 1826)	F	0	2	0	3	1	0	2	0	0	2	1	1	2	0	0	4
<i>Pseudamussium clavatum</i> (Poli, 1795)		1	0	0	1	0	0	1	0	0	0	0	0	0	0	0	1
Pectinidae (D)		0	2	2	1	1	0	2	1	1	2	1	3	2	1	5	2
Anomiidae ( <i>Monia patelliformis</i> (Linnaeus, 1761))	F																
<i>Heteranomia squamula</i> (Linnaeus, 1758))		4	4	10	4	7	5	7	6	5	6	10	10	9	7	8	14
<i>Abra prismatica</i> (Montagu, 1808)	D	9	10	5	9	13	7	4	14	7	7	6	8	9	3	11	11
<i>Centrocardia aculeata</i> (Poli, 1795)		0	0	1	2	0	0	0	0	0	0	0	1	0	0	1	0
<i>Parvicardium minimum</i> (Philippi, 1836)	F	58	78	49	61	77	49	57	69	47	42	40	49	56	51	133	143
<i>Timoclea ovata</i> (Pennant, 1777)	F	49	75	61	52	53	36	33	47	35	32	42	63	47	38	105	131
<i>Cardiomya costellata</i> (Deshayes, 1835)		1	2	0	1	1	0	1	0	1	0	3	0	2	1	5	1
<i>Cuspidaria cuspidata</i> (Olivi, 1792)		0	0	0	1	0	0	1	0	0	1	0	0	2	0	1	1
<i>Myrtea spinifera</i> (Montagu, 1803)	S	2	3	4	2	1	2	2	3	3	3	2	1	4	2	6	3
<i>Clausinella fasciata</i> (da Costa 1778)	F	7	6	1	8	8	6	7	4	5	2	3	2	6	6	13	15
<i>Corbula gibba</i> (Olivi, 1792)	F	4	3	0	1	4	5	4	2	3	3	6	2	4	7	9	8
<i>Hiatella arctica</i> (Linnaeus, 1767)	F	7	20	11	14	14	17	15	17	12	7	12	14	9	14	24	33

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Table 2 (continued)

CV0518-MC2-T1	L	0-1 cm	1-2 cm	2-3 cm	3-4 cm	4-5 cm	5-6 cm	6-7 cm	7-8 cm	8-9 cm	9-10 cm	10-11 cm	11-12 cm	12-13 cm	13-14 cm	14-15 cm	15-16 cm
87,3 m	WW	106.35	104.65	83.24	90.30	91.96	99.64	102.91	88.16	94.62	88.97	102.26	99.98	107.64	88.42	101.43	128.01
Veneridae		19	33	19	25	16	14	14	16	20	7	21	13	21	17	31	40
<b>Gastropoda</b>																	
<i>Calyptraea chinensis</i> (Linnaeus, 1758)	F	14	11	6	12	7	6	7	7	7	3	7	7	6	3	14	12
<i>Lepetella espinosae</i> Dantart & Luque, 1994		0	2	0	3	0	0	0	1	0	0	1	1	1	0	1	1
<i>Diodora</i> sp.		1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Buccinidae		0	0	0	0	0	0	0	2	0	0	1	0	1	0	0	1
<i>Caliostoma</i> sp.	C	1	3	2	2	2	2	2	2	2	0	3	1	0	1	2	4
<i>Turritella communis</i> Risso, 1826	F	14	17	10	18	7	11	9	7	6	12	11	11	18	18	22	30
<i>Bittium submamillatum</i> (Rayneval & Ponzi, 1854)		12	14	12	16	8	15	11	4	12	15	7	14	13	15	13	14
<i>Eulima bilineata</i> Alder, 1848	C	1	0	2	0	2	1	2	1	1	0	2	1	2	2	2	5
<i>Alvania</i> spp. ( <i>Alvania punctura</i> (Montagu, 1803) -	D																
- <i>Alvania testae</i> (Aradas & Maggiore, 1844))		1	0	0	2	0	2	2	3	2	1	2	1	1	1	0	1
<i>Hyalia vitrea</i> (Montagu, 1803)	D	2	0	1	3	2	0	2	5	2	3	7	2	4	3	3	4
Rissoidae	D	0	0	1	0	0	2	0	1	0	1	1	1	1	1	1	4
<i>Aporrhais pespelecani</i> (Linnaeus, 1758)		0	0	0	0	1	1	2	0	0	0	1	0	0	0	0	1
<i>Capulus ungaricus</i> (Linnaeus, 1758)		0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0
Patellidae		0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	1
Naticidae	C	0	3	0	1	1	0	1	0	2	0	2	2	1	1	4	3
<i>Bolma rugosa</i> (Linnaeus, 1758)		0	0	1	0	0	0	0	0	0	1	0	0	0	0	1	0
<i>Trophonopsis muricata</i> (Montagu, 1803)		0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0
<i>Euspira nitida</i> (Donovan, 1804)		0	0	0	0	0	0	0	0	1	0	0	0	0	0	1	0
<i>Fusinus</i> sp.	C	2	1	0	0	2	1	1	0	1	0	1	0	3	1	2	0
<i>Mangelia costata</i> (Pennant, 1777)	C	3	2	1	0	2	2	3	2	1	3	1	1	2	4	1	2
<i>Sorgenfreispira brachystoma</i> (Philippi, 1844)	C	5	3	1	3	7	5	5	5	5	3	2	4	6	5	5	8
<i>Turbonilla sinuosa</i> (Jeffreys, 1884)		1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
<i>Eulimella</i> sp.		1	0	0	0	1	1	0	0	0	0	1	1	0	1	1	1
<i>Weinkauffia turgidula</i> (Forbes, 1844)		2	0	0	0	0	0	0	1	0	0	0	0	1	0	0	0
<i>Acteon</i> sp.	C	0	0	0	2	0	1	0	1	0	0	1	1	0	0	3	1
Bullidae	C	0	0	0	2	2	2	2	2	2	2	1	0	1	1	1	3
<i>Cylichna cylindracea</i> (Pennant, 1777)	C	1	2	0	3	4	3	3	0	2	1	3	2	1	1	0	1
Scaphandridae		0	0	0	0	0	0	2	0	2	1	1	0	2	0	1	2
<i>Hermania scabra</i> (O.F. Müller, 1784)		0	0	0	0	0	2	0	0	1	1	0	0	0	0	1	1
<b>Scaphopoda</b>																	
<i>Antalis</i> spp.		8	0	0	17	11	16	19	14	11	12	14	12	17	11	0	22
<i>Entalina tetragona</i> (Brocchi, 1814)		1	0	0	2	1	0	2	0	1	0	0	0	0	0	0	0
<i>Cadulus</i> sp.		0	0	0	0	2	0	0	0	0	0	0	0	0	0	0	0
<b>Pteropoda</b>																	
<i>Cavolinia inflexa</i> (Lesueur, 1813)		17	58	43	29	33	25	50	28	25	11	36	37	39	27	42	45
<i>Clio pyramidata</i> Linnaeus 1767		3	0	11	9	4	8	9	3	14	3	2	7	2	6	1	6
<i>Creseis acicula</i> (Rang, 1828)	110	112	103	135	76	83	112	33	56	54	70	56	34	41	59	44	
<i>Limacina inflata</i> (d'Orbigny, 1834)		9	1	8	19	4	11	20	18	10	28	16	25	7	24	3	11
<i>Peraclis reticulata</i> (d'Orbigny, 1835)		0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0
<b>Caenogastropoda</b> ( <i>Atlanta</i> sp.)		1	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0
<b>Cephalopoda</b> ( beaks)		0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	1
<b>Echinodermata</b> ( <i>Cidaris</i> sp.)		0	0	2	0	0	2	0	0	0	0	0	0	1	0	1	1
<b>Cnidaria</b>																	
Hydrozoa		0	3	2	2	2	1	1	1	0	0	3	0	0	0	0	0
<i>Stephanoscyphus</i> sp.		0	1	3	2	1	0	1	3	3	2	4	0	0	0	0	0
Caryophyllidae		1	1	0	1	0	1	2	1	0	0	1	1	1	0	1	1
<b>Pisces</b>																	
<i>Engraulis encrasicolus</i> (Linnaeus, 1758)		0	0	0	0	0	0	0	1	0	0	0	0	0	1	0	0

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Table 2 (continued)

CV0518-MC2-T1	L	0-1 cm	1-2 cm	2-3 cm	3-4 cm	4-5 cm	5-6 cm	6-7 cm	7-8 cm	8-9 cm	9-10 cm	10-11 cm	11-12 cm	12-13 cm	13-14 cm	14-15 cm	15-16 cm
87,3 m	WW	106.35	104.65	83.24	90.30	91.96	99.64	102.91	88.16	94.62	88.97	102.26	99.98	107.64	88.42	101.43	128.01
<i>Sardinella aurita</i> Valenciennes, 1847		0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0
<i>Seriola</i> spp. (scales)		0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0
<i>Trachurus</i> sp.		1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
<i>Cepola macrophthalma</i> (Linnaeus, 1758)		0	0	0	0	0	0	1	1	0	0	0	0	0	0	0	0
<i>Merluccius merluccius</i> (Linnaeus, 1758)		0	0	0	0	0	0	0	0	0	0	0	0	0	0	2	0
<i>Micromessistius poutassou</i> (Risso, 1827)		0	0	0	0	0	1	0	0	0	0	0	0	0	0	1	0
Gadiforme ( <i>Trisopterus</i> ?)		0	1	0	0	0	0	0	1	0	0	0	0	0	0	0	0
<i>Benthoosema glaciare</i> (Reinhardt, 1837)		1	0	0	0	0	1	0	0	0	0	1	0	1	0	0	0
<i>Ceratospopelus maderensis</i> (Lowe, 1839)		0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1
<i>Maurolicus mueleri</i> (Gmelin, 1789)		0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	1
Stomiidae ( <i>Chauliodus sloani</i> Bloch & Schneider, 1801)	0	0	0	1	0	0	0	0	0	0	0	0	1	0	0	0	0
<i>Buenia</i> sp.		0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0
<i>Callionymus</i> sp.		0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0
<i>Chromogobius zebratus</i> Miller, 1971		0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0
<i>Deltentosteus quadrimaculatus</i> (Valenciennes, 1837)	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0
<i>Gobius</i> sp. (Gobiidae)		0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0
<i>Lesuerigobius friesi</i> (Malm, 1874)		0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	1
<i>Pomatochistus</i> sp. (Gobiidae)		2	1	1	1	1	2	0	1	4	2	2	3	2	0	1	0
<i>Coris julis</i> (Linnaeus, 1758)		0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0
Ophidiiforme ( <i>Carapus acus</i> Brännich, 1768)		0	0	0	0	2	0	1	0	1	1	0	0	0	0	0	0
<i>Stromateus fiatola</i> Linnaeus, 1758		0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0
<i>Conger conger</i> (Linnaeus, 1758)		0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	1
<i>Arnoglossus</i> sp.		0	0	0	0	0	1	0	0	0	0	1	0	0	0	0	0
<i>Symphurus</i> sp. (scale)		0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1
Fish ossicle		3	0	0	1	0	1	0	0	1	1	0	1	2	0	0	0
Unidentified otholit		0	0	0	0	0	0	0	0	1	0	0	0	2	0	0	0
<b>Other remains</b>																	
Wood remains		0	3	2	0	0	0	2	1	3	1	1	0	3	1	3	0
Vegetal fibers (Marine Phanerogams ?)		2	1	0	0	0	0	0	1	0	0	2	0	1	0	1	0
Microplastics (balls)		1	3	1	0	1	3	0	0	0	0	0	0	0	0	0	0

3.3.2. Dead fauna at MC2: Abundance in the sediment core

A total of 7383 items (basically faunal remains) were sorted and classified in MC2. The resulting data matrix had 113 taxa/categories. The most diverse compartment (dead fauna) was the benthos, with 28 bivalve genera/species, 23 gastropods, 20 teleosteans, and 6 decapod crustaceans in decreasing order. Mesopelagic fauna were represented by pteropods, with 6 species, and scarce individuals of some meso- and epipelagic fish (4 myctophids and 3 epipelagic species: *Engraulis encrasicolus*, *Sardinella aurita*, and *Trachurus* sp.).

3.3.2.1. Zooplankton species. Pteropods (pelagic zooplankton) exhibited important changes depending on the species (Fig. 5). *Creseis acicula* shells were less abundant at deeper (older) levels of the core below 8 cm (range of abundance: 31.3–68.4 ind./100 g sediment), increasing significantly (1-way ANOVA  $F_{2,13} = 11.5$ , Fisher test comparing 0–7 vs 8–13,  $p < 0.001$ ) at levels between 0 and 7 cm and 82.9–142.9 ind./100 g sediment, with a peak at 4 cm decreasing towards the surface (103.3 ind./100 g sediment). *Cavolinia inflexa* showed an intermediate abundance range (12.6–55.4 ind./100 g) in all cores, without significant differences among levels; there was some decrease observed from the deepest level (at 16 cm, 35.3 ind./100 g) to 11 cm (12.6 ind./100 g), with an increase to the surface (1–2 cm, 55.4 ind./100 g). *Clio pyramidata* had nearly similar shell abundances to those of *C. inflexa*, with lower values at intermediate levels (at 10–11 cm, 2–3.4 ind./100 g) and in the first 0- to 2-cm surficial layer (0–2.7 ind., Fig. 5), but the results were always low. Limacinids were not plotted because no evident tendencies were identified.

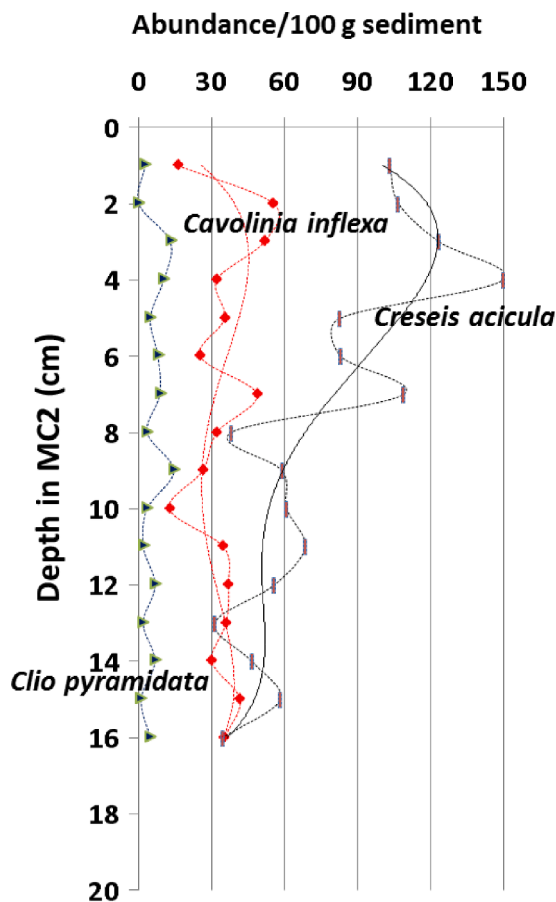


Fig. 5. Standardized abundances (number of individuals/100 g sediment) of dominant taxa found in sediment core MC2 against depth in the core (cm) for the main 3 pteropod species identified (*Clio pyramidata*, *Cavolinia inflexa*, and *Creseis acicula*). The corresponding ages (years) of sediments, calculated by the CSR model, are shown in Table 1.

3.3.2.2. Benthos: Sessile fauna (corals). Quantification of sessile benthic fauna was possible in MC2 for corals (mainly Octocorallia). In all sediments down the whole MC2 core, we found sclerites (the structures covering coral colonies) of corals, including soft corals (Alcyonaria), sea pens (Pennatulacea) and other (i.e., the rest of) Octocorallia. From the surface to the bottom (Fig. 6), the sclerite density ranged between 51.6 sclerites/100 g sediment at the surface (0–1 cm) to a maximum value throughout the MC2 of 207.3 sclerites/100 g sediment, measured at the 5- to 6-cm level. We found a decrease in sclerites with depth (with fluctuations), with measured values ranging between 149.4 sclerites/100 g of sediment at 9–10 cm and a minimum of 28.7 sclerites/100 g of sediment at 12–13 cm (Fig. 6). The increase in sclerites since the date at which the area was declared a marine reserve (in the 1980 s, between the 5 and 6 cm in MC2) was not significant (1-way ANOVA  $F_{(2,13)} = 2.9$ ,  $p = 0.08$ ; Fisher LDS  $p = 0.05$  comparing levels 0–5 cm and 6–10 cm). The three levels corresponded to the periods 2017–1989 (0–5 cm), 1989-ca. 1927 (6–10 cm) and older than 1927 (>11 cm) under the assumptions of the CRS dating model (Table 1). The 0- to 5-cm layer comprised the period of the marine reserve declaration, the 6- to 10-cm layer characterized the period with the highest fishing activity on the shelf, and the older (>11 cm) layer was a period of no or negligible fishing activity. The mean numbers of sclerites for the 3 periods were  $164.3 \pm 60.2$  (C.I. 95 %) sclerites/100 g,  $88.7 \pm 42.4$  sclerites/100 g and  $94.2 \pm 34.8$  sclerites/100 g, from the shallower to deeper layers, respectively.

3.3.2.3. Benthos: Bivalves and gastropods (abundance of dominant species).. Regarding the most abundant species in the MC2 profile, we found the same general tendency, a “U-shaped” model with the lowest densities at intermediate levels of MC2, ca. the 6–9(10) cm layers, the period 1980-ca. 1927 with the highest fishing activity. Among bivalves, the two (filter-feeder) *Cardiidae* *Timoclea ovata* and *Parvicardium*

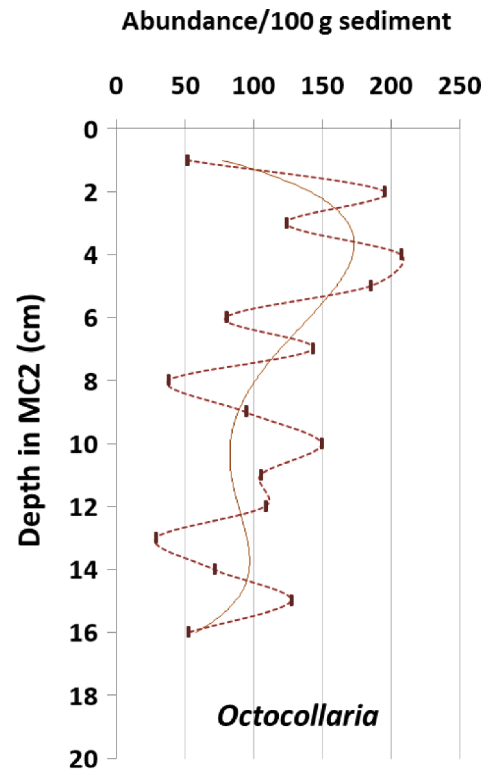


Fig. 6. Standardized number of sclerites/100 g sediment for corals (Octocorallia) representing the sessile fauna in the sediment core of MC2 against depth in the core (cm). The corresponding ages (years) of sediments calculated by the CSR model are shown in Table 1.

*minimum* had parallel trends in their abundances in the MC2 profile, with the highest densities measured at the deepest levels (>14 cm), similarly low densities measured from 6 to 14 cm and increases in most surface levels of the MC2 profile ( $F_{(2,13)} = 2.8$ ,  $p = 0.09$  for *T. ovata*, significant Fisher  $p = 0.03$  comparing 0–5 cm vs 6–10 cm; *P. minimum* no-significant results, Fig. 7). *Saccella commutata* and *Hiatella arctica* showed the same common trend, with the highest abundance values recorded at the deep and surface layers of the MC2 profile and the lowest abundance values measured at intermediate levels (Fig. 7), though these tendencies were not significant (at 0–5 cm). Among gastropods, we found the same “U” tendency than for bivalves for the most abundant species, *Turritella communis*, and to a lesser extent for another dominant species, *Calyptraea chinensis* (Fig. 7), both filter feeders. In both examples, with a significant increase at 0–5 cm ( $F_{(2,13)} = 4.4$  and 2.5, respectively; Fisher  $p = 0.01$  and 0.04, respectively) compared to the 6–10 cm period.

3.4. Benthos diversity (whole community; top predators: Fish and crustaceans)

Similar levels of diversity ( $H = 2.5$ ) were observed in all MC2 profiles for the whole (benthic) community (Fig. 8). This is an expected result considering the high permanency of benthos communities, wherein species can change but diversity remains more or less at a constant level. However, two shifts in diversity were observed at 4 cm (1992–1998) and 11 cm (ca. at the beginning of the last century), which may represent changes in benthic succession. At the most surficial level (last decade), there was a decrease in  $\dot{H}$  (Fig. 8).

Fish were not highly abundant in the MC2 sediments (69 individuals counted), although they were moderately diversified (23 species), being benthic-species dominant, and the small Gobiidae *Pomatoschistus* sp. (27.5 %) was the most abundant species identified. Together with decapod crustaceans, benthic fish represent the top predator scavengers in MC2. At 11–16 cm, i.e., beyond SXX, the diversity levels were low ( $\dot{H}=1.5$ ), increasing between 10 and 3 cm to  $\dot{H}=2$ . In the most surficial levels of the MC2 profile, since ca. 2000,  $\dot{H}$  decreased again (Fig. 8).

3.5. Community analysis (dead fauna)

3.5.1. Inertial analysis

Multidimensional scaling (MDS) for MC2 showed some distributional tendencies among the analysed 1-cm levels (Fig. 9) along the inertial axes. The MDS analysis also showed tendencies in the distribution of the 4 groups of samples considered (Fig. 9), based on the entire

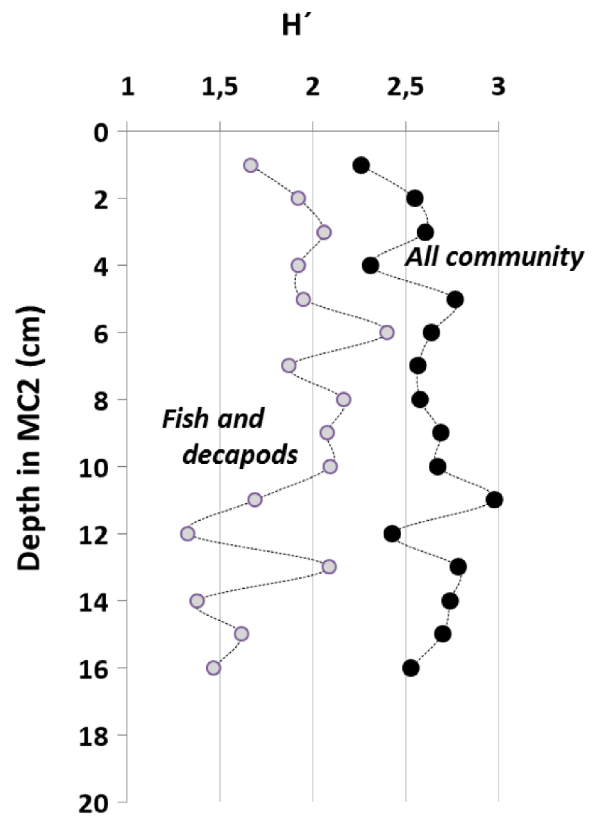


Fig. 8. Diversity (Shannon index,  $\dot{H}$ ) of the whole benthic community and of top predator scavengers in the sediment core MC2 against depth in the core (cm). The ages (years) of sediments calculated by the CSR model is shown in Table 1.

community, when analysed by separate *epi*- and mesopelagic species and benthic-epibenthic invertebrates.

For zooplankton (*epi*- and mesopelagic species, Fig. 9a), groups of samples were distributed in the plot from the most surficial levels on the right to the deepest levels in the core on the left, with few exceptions (0–1 cm, 14–15 cm). The deepest levels (11–16 cm), which would approximately correspond to levels of > 110 years before the current year of sampling (2018), were also well-seggregated. The factor “core level” showed almost significant differences in the main PERMANOVA

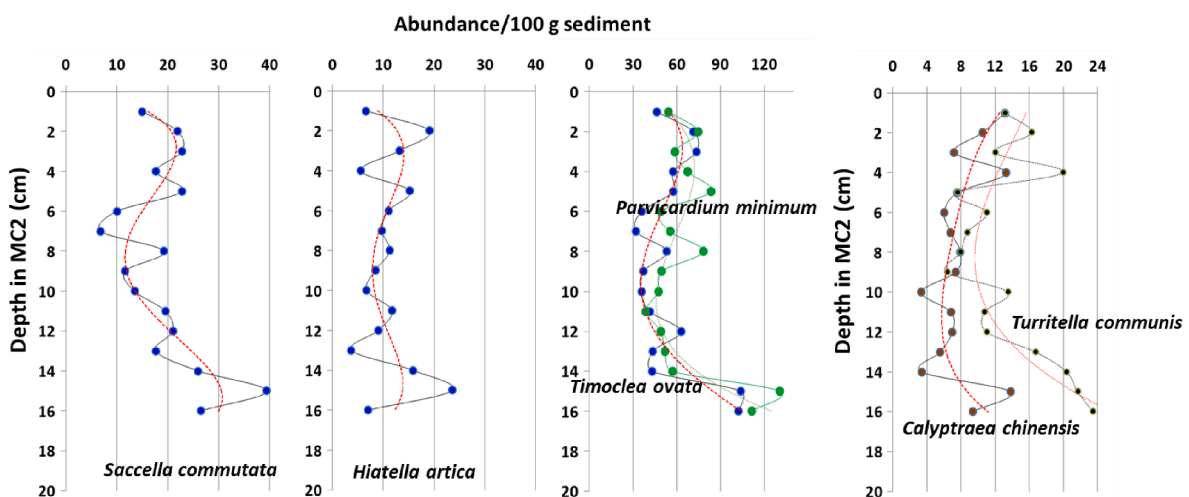


Fig. 7. Standardized abundances (number individuals/100 g sediment) of the four dominant bivalves and 2 gastropods (left) in the sediment core MC2 against depth in the core (cm). The corresponding ages (years) of sediments calculated by the CSR model are shown in Table 1.

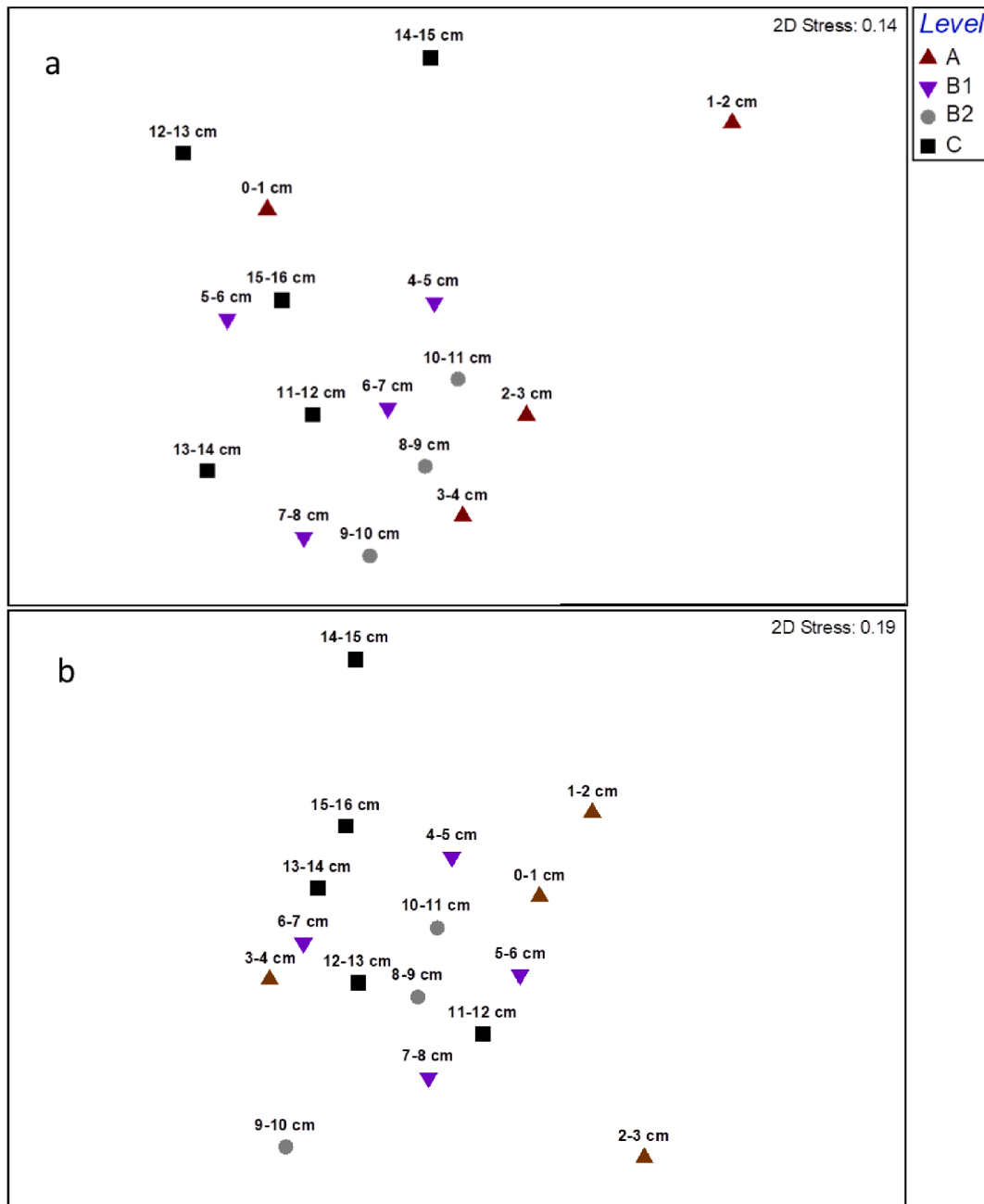


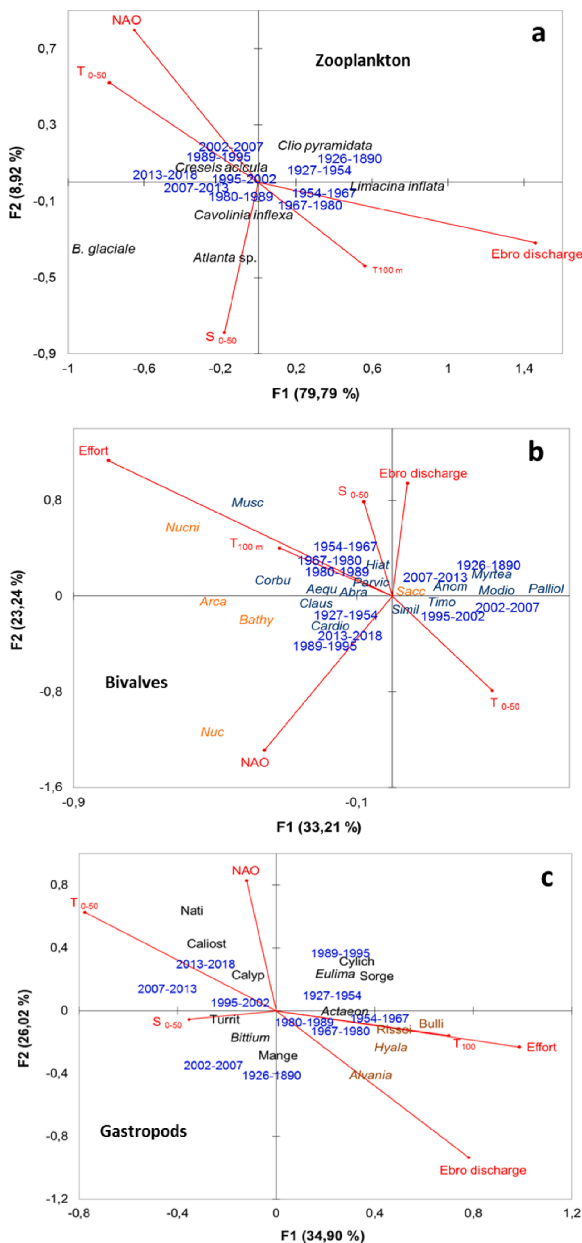
Fig. 9. Multidimensional scaling (MDS) analysis of the whole community of reconstructed (dead) fauna, based on the MC2 analysed every 1-cm level, with the 4 groups established (see methods for details): A (0–1 to 3–4 cm); B1 (4–5 to 7–8 cm); B2 (8–9 to 10–11 cm) and C (11–12 to 15–16 cm). The analysis was performed on a) meso- and benthopelagic fauna (mainly pteropods) and b) on the entire community of benthos (mainly invertebrates).

test ( $t = 1.72$ ;  $p = 0.07$ ), and significant in the pairwise tests between the A and C groups (i.e., 0–4 cm vs 11–16 cm;  $t = 1.60$ ;  $p = 0.04$ ) and between the B2 and C groups (8–11 cm vs 11–16 cm;  $t = 1.92$ ;  $p = 0.03$ ), the latter of which constitutes a comparison of two contiguous groups. The benthos groups (Fig. 9b) were also distributed in the plot from the right (surface levels at MC2) to the left (deepest levels at MC2), though any test (main and comparison between sample groups) was significant ( $t_{\text{main}} = 1.21$ ;  $p = 0.14$ ).

### 3.5.2. Environmental analysis

The CCA results based on the MC2 profile showed some relationships among the dominant species in communities and environmental variables (Fig. 10). The CCA performed on zooplankton (basically pteropods) explained 88.7 % of the variance in the two first axes. Although

the Permutation test, giving the significance of the relationship between the contingency table of species/abundance and the explanatory variables, was not significant ( $p > 0.05$ ), positive NAO periods (low Ebro River discharges), which were also related to higher surface (0–50 m) temperatures, were positively related to *Creseis acicula* abundance (especially in the period 1980–2017, Fig. 10a). The CCA performed on epibenthic predators (fish, decapods, mainly) explained 69.3 % of the variance in the two first axes. Any clear relationships were identified among the species and variables, and the fishery effort was not important (had no influence on species) as an explanatory variable (Figure S1). Permutation test was not significant in these CCA ( $p > 0.05$ ). The CCA performed on bivalves (benthos) explained 56.4 % of the variance in the two first axes. Permutation test was significant for this CCA (Pseudo  $F = 1.7$ ;  $p = 0.01$ ). Fishery effort was the main explanatory



**Fig. 10.** Dominant species abundances (remains) in MC2 distributed on the main CCA axes, relating those taxa (species) identified in the sediment core with environmental variables. The numbers indicate the years representing the periods of years listed in Table 1, which are associated with each 1-cm slice of sediments dating back to ca. 110 years ago.  $T_{0.50\text{ m}}$  is the mean temperature of the surface water column to a depth of 50 m, and  $T_{100\text{ m}}$  is the mean T at the nearest depth of the MC2 sampling site (100 m), with the same notation used for salinity (S). The NAO is the annual North Atlantic Oscillation climatic index, and effort represents the mean number of boats in Castelló (the nearest port to Columbretes) within each period. Effort was not considered in the zooplankton analysis. CCAs were performed for the zooplankton (a) and benthos communities: bivalves (b); gastropods (c). Some species in CCA represented by abbreviations of generic/Family names. Bivalves: Nucni: *Nucula nitidosa*; Bathy (*Bathyarca* sp.); Musc (*Musculus subpictus*); Nuc: *Nucula handleyi*; Corbu: *Corbula gibba*; Aequi (*Aequipecten* sp.); Claus (*Clausinella fasciata*); Cardio (*Cardiomya costellata*); Hiatt (*Hiattella arctica*); Sacc (*Saccella commutata*); Parvic (*Parvicardium* sp.); Simil (*Similipecten* sp.); Anom (Anomiidae); Timo (*Timoclea ovata*); Modio (*Modiolus phaseolina*); Palliol (*Palliolum incomparabile*). Gastropods: Nati (Naticidae); Calio (Caliostomidae); Turrit (*Turritella communis*); Calyp (*Calyptrea chinensis*); Mange (*Mangelia costata*); Cylich (*Cylichna cylindracea*); Sorge (*Sogenfreispira brachystoma*); Bulli (Bullidae); Rissoi (Rissoiidae, undetermined).

variable, with filter feeders such as Pectinidae (*Palliolum* sp. showed this tendency more clearly) and other species (*Modiolus* spp., *Anomia* sp. and *Timoclea ovata*) dominating when fishery efforts decreased, being distributed towards the right side in Fig. 10b. *Myrtea spinifera*, which also appears with filter feeders, belongs to a chemosymbiotic family characterized by bearing symbiont bacteria helping in C fixation and  $N_2$  fixation. In contrast, Protobranchia (the clearest example, *Nucula nitidosa*), a deposit feeder-infauna, increased its abundance under higher fishery efforts (towards the left side in Fig. 10c) linked to the period from 1954 to 1989, during which the number of boats operating at the nearest port considered was the highest. Among gastropods (Fig. 10c), we found a similar tendency though permutation test for CCA was not significant, Pseudo F = 1.3;  $p = 0.20$ ). As for bivalves, few deposit feeders (Rissoiidae, *Alvania* spp.; Iravadiidae, *Hyala vitrea*), all considered subsurface deposit feeders, were more abundant with higher fishery effort (and river discharge) conditions. Under opposite conditions, suspension feeders (*Calyptrea chinensis*, partially *Turritella communis*) and carnivores (e.g., Naticidae, *Caliostoma* sp., Mangelidae, *Actaeon* sp.) increased. Deposit feeders were linked with the period 1954–1989, exactly as reported for bivalves. Schematically, for those species of bivalves that occupied extreme positions in the CCA (*Nucula* spp. vs *Palliolum* spp.). There was an increase (recovery) in filter feeders (FF) vs deposit feeders (DF) (Fig. 4) with a decrease in effort (n° boats in the area) after the Reserve declaration in 1987–1990.

#### 4. Discussion

##### 4.1. Dating and validation of sediment core

The main geochronological result derived from radionuclide analyses is that the sediment 11 cm below the surface, where  $^{210}\text{Pb}_{\text{xs}}$  disappears, separates the sediments that are older and younger than ca. 110 years (ca. 1908). The mass accumulation rate (MAR) measured from the  $^{210}\text{Pb}$  profile of MC2 in the first 0–6 cm (to the 1980 s) was  $0.170\text{ g cm}^{-2}\text{ y}^{-1}$  based on a constant rate of supply (CRS) model (Appleby and Oldfield, 1978). Our  $^{210}\text{Pb}_{\text{xs}}$  dating was validated by different changes in Hg analyses, i.e., peaks or increases/decreases associated with concrete events and by microplastic appearance in the core. This dating is within the standards of core dating compared with other paleobiological studies (see Table S1), performed in oldest periods in the Mediterranean. First, the CRS model was applied by different authors (see Table S1). Furthermore, in previous approaches (details in Table S1), we found coarser core slicing, higher  $^{210}\text{Pb}$  uncertainties (see Schirone et al., 2022) and, when radiocarbon was analysed to validate  $^{210}\text{Pb}$  dating for recent (last 100 yrs) samples, highly variable radiocarbon results, even after they were corrected for the “marine reservoir effect”. Additionally, similar MAR estimates to ours have been found over the Catalanian shelf at comparable depths of 70–80 m ( $0.18\text{ g cm}^{-2}\text{ y}^{-1}$ , Sánchez-Cabeza et al., 1999). However, when MARs were measured at shelf stations in the Gulf of Lyons, close to the influence of the Rhône River (Miralles et al., 2006), the results were clearly higher (between 0.33 and  $7.0\text{ g cm}^{-2}\text{ y}^{-1}$ ) than those off the Columbretes Islands. In the MC2 station at Columbretes area, ca. 95 km from the Ebro River mouth, only some indirect influence of the Ebro River plume could be recorded. After the Columbretes Islands were declared a protected area (in 1987–1990), trawling was forbidden, and fishery efforts were reduced to traditional nondestructive activities. The  $^{210}\text{Pb}$  profile of MC2 became situated the period at which the cessation of such activities occurred in the 1980 s (1980–1989), at 5–6 cm.

Other human activities impacted the Columbretes ecosystem. The Carallot (Bergantin) islet, which is close to station MC2, was used by the U.S. Army for seven years as a target for firing training grounds for plane pilots until 1982, when military manoeuvres were finished. In sediments collected from military grounds, mercury (Hg) concentrations can be higher because this metal has been used as an ammunition primer and detonator (Gębka et al., 2016). The Hg analyses performed at MC2



evidenced significant mercury peaks at 6–7 and 5–6 cm, which correspond to the periods of 1967–1980 and 1980–1989, validating radiocarbon dating and coinciding with the period of military activities in Columbretes and decreasing after the activities stopped in 1982. Pollution by Hg can also originate from industrial activities (Sánchez-Cabeza et al., 1993; Armenteros et al., 2016), and the observed increase in mercury in MC2 sediments above 10 cm (ca. the beginning of the SXX century) coincided with the beginning of industrialization in NE Spain, which was reflected in sediments collected off the central coast of Catalonia (off Barcelona: Sánchez-Cabeza et al., 1993; off Vilanova: current RECOMARES data, authors unpublished), ca. 160–200 km N of Columbretes. This Hg increase would be related to local industry in the area, even with the cited industrial activity off the Catalonia coasts, since the general current regime in the area trends from NW to SE (Font et al., 1988). Finally, the uppermost (in the 0–1 cm layer) decline in Hg observed in the MC2 profile could indicate the success of recent directives, such as the Marine Strategy Framework Directive, for reaching a good environmental status. A similar decrease in Hg has been observed in mussels collected close to Columbretes (Peñíscola) in recent decades (from 0.3 mg/kg dry weight in the early 1990 s to 0.1 mg/kg in 2013, Campillo et al., 2019).

A final time marker was supplied by microplastics, which were represented by balls of fibres in the MC2, as also found in the stomach contents of deep-water crustaceans (Carreras-Colom et al., 2018) and in sediments (Carreras-Colom et al., 2020) in the Balearic Basin. Fibre balls were recognized in the MC2 profile up to the 6- to 7-cm layer, corresponding to the period 1967–1980. Industrial production of plastics began in the 1950 s, and the first findings of these materials in marine sediments occurred in the early 1970 s (Carpenter and Smith, 1972; GESAMP, 2015) and until 1987 in the study area (Carreras-Colom et al., 2020), but not before because microplastics were not analysed until recent years. In conclusion, the two independent markers used (Hg and microplastics) confirm the  $^{210}\text{Pb}_{\text{xs}}$  dating of the MC2 profile.

## 4.2. Analysis of communities

### 4.2.1. Possible bioturbation and fauna mixing

Bioturbation is considered a possible source of “faunal mixing” at the uppermost levels of cores. The mixing level in MC2 (the 0–4 cm level, where we found > 90 % of the living fauna) coincided with the mixing level resulting from bioturbation recorded in the upper slope/shelf (4 cm at station CN-32, Sánchez-Cabeza et al., 1999) off the central Catalan coasts. The slightly higher % water at 0–4 cm of MC2 sediments (26.9–29.3 %) compared to those of deeper levels (<23 %) suggested a low bioturbation by living fauna, which seems logical since all belonged to small meiofauna, reaching low densities (1.5 ind/10 cm<sup>2</sup> for nematods). At MC2, practically all benthic molluscs found were small, as small as foraminiferans, often. We found some consistent tendencies, e.g., the link between benthic bivalve/gastropod feeding guilds and different levels of fishing effort (see below), in our analysis, suggesting that the real age of the shells of the small benthic molluscs counted matched the estimated age intervals for the sediments where they were immersed. A known limitation when shells are counted as “valid” is that some might be reworked and can be significantly older than the dating of the sediment layers from which these shells have been taken. This can be in part corrected when only entire shells are counted (see Armenteros et al., 2016; Table S1). In this way, the use of radiocarbon ( $^{14}\text{C}$ ) dating on shells was used recently to validate  $^{210}\text{Pb}$  profiles (Tomašových et al., 2017). These radiocarbon dating methods have been considered with caution due to the marine reservoir effect, which may also cause age overestimation, with this effect being higher due to outcrops of older carbonates found in open/deep waters (Mangerud, 1972; see also Table S1). In fact, studies on recent sedimentation rates (last ca. 100 yrs, Sánchez-Cabeza et al., 1993, 2012; Miralles et al., 2006) were not based on radiocarbon but on the  $^{210}\text{Pb}$  approach adopted here. The radiocarbon analysis of young material is often associated with large

uncertainties (Mangerud, 1972; Preusser and Schreurs, 2011). Even so, modern studies applied corrections to the marine reservoir effect for radiocarbon (Vidovic et al., 2016; Gallzemer et al., 2021). Although the results, after a collection of assumptions and corrections, continue showing high associated errors due to (median) age estimates (Vidovic et al., 2016), and the validation of C14 vs  $^{210}\text{Pb}$  profiles is based on few observations (Tomašových et al., 2017, Fig. 1C, see S1 Table), some authors concluded that entire shells collected at core modern layers (last 100 yrs) are also recent. This would validate our results, assuming that the shells counted correspond to the same period that the sediment slices from which these shells were taken.

The continuous shape of the  $^{210}\text{Pb}_{\text{xs}}$  profile of MC2, allowing calculations of sedimentation rates with the CRS model, indirectly suggests the absence of catastrophic events affecting sedimentation. This would reduce a massive mix of faunal remains of different sediment levels or the deposition of remains from neighbouring areas, at least in the upper part of MC2 (to the level of the  $^{210}\text{Pb}_{\text{xs}}$  extinction at 11 cm). Furthermore, all the species identified in MC2 belong to the current outer shelf communities of benthic molluscs, which are excluded in the MC2 species from other depths or areas (e.g., absence of invasive species).

### 4.2.2. Representativeness of the core MC2

Few studies conducted in shelf communities have attempted recent reconstructions (ca. over the last 100 years) of macrofaunal communities while simultaneously considering the chronology of sediments and analyses of the dead fauna (Armenteros et al., 2016). The reconstruction done on the MC2 station can likely be extrapolated to the soft “muddy or sandy-muddy infralittoral and circalittoral assemblages” in the Columbretes MPA, one of the widest distributed assemblages in the reserve to depths of 95 m (ERENA, 2014). Additionally, the community of bivalves and gastropods of MC2 represented the current typical outer continental shelf communities of benthic molluscs living in the Iberian region (Gofas et al., 2017). Our previous experience off the Ebro Delta deep-sea region (MC4, 398 m depth) in the Balearic Basin evidenced important and interpretable long-term changes due to zooplankton (pteropods) oscillations (Cartes et al., 2017, 2018). The same increase in the pteropod *Creseis acicula* was associated with warming periods that occurred at the MC2 station off the Columbretes shelf. *Creseis acicula* increased at most surface levels not only of MC2 but also at the same surface levels of MC5, a core also collected from the outer shelf, 12 km to the E of MC2, in a fishing area out of the MPA (see Fig. 1, map). Therefore, some results derived from the MC2 reconstruction seem extrapolable to neighbouring areas to the Marine Reserve.

### 4.2.3. Chronology of biological changes and faunal diversity

In our chronological analysis at MC2, we identified three periods: i) a first, modern level (from the present day to 1980) at 0–6 cm with a similar MAR range (0.21–0.12 g cm<sup>-2</sup> y<sup>-1</sup>); ii) an intermediate level spanning from 1980 to ca. 1908 (i.e., 110 years ago) between 6 and 11 cm; and iii) sediments below 11 cm, corresponding to periods before SXX. Most species analysed, the clearest example of the most abundant bivalves and gastropods, followed a “U-shaped” abundance pattern in MC2, with a decrease in abundance observed ca. 9–11 cm, and a later, more modern increase at 5–6 cm (1980–1989), mainly significant among filter-feeder bivalves/gastropods. This increase characterizes the period in which trawling was forbidden in the Columbretes Reserve. Such changes in benthic bivalves/gastropods were also evidenced from a trophic perspective; as the species changed from more (subsurface) deposit feeders (DF: *Nucula nitidosa*, *Batharca*, *Hyala*...), which were more abundant under higher trawling efforts, to filter feeders (FF, e.g., Pectinidae among bivalves) after the cessation of trawling, i.e., in the conditions generated by the establishment of the marine reserve. Species, however, can show opportunistic strategies (Cadée, 1984), and a possible relationship between feeding guilds and human impact cannot be so directly established. Among bivalves, *Corbula gibba* (a filter feeder but also a deposit raker) has an opportunistic strategy that allows it to

quickly colonize disturbed communities (Hrs-Brenko, 2006), such as in conditions found under high fishing activity (as evidenced in the CCA). *Bathyrca* can be both a surface deposit and a nonselective filter feeder (Renaud et al. 2011). Even so, the significant increase of some filter-feeder bivalves (*Timoclea ovata* and *Parvicardium minimum*), and gastropods (*Turritella communis* and *Calyptrea chinensis*), together with changes in the ratio between DF/FF (schematized in Fig. 4) evidenced how there was an increase (recovery) of filter feeders with a decrease in effort (n° boats in the area) after the Reserve declaration at the end of the 1980s (1987–1990). Changes in the relationship between deposit and filter feeders have been evidenced in studies on trawling impact (Allen and Clarke, 2007; de Juan et al., 2007). We evidenced this at a long-term scale after MPA creation. An increase in deposit feeders can also be related to natural increases in river discharge, as discussed by Armenteros et al. (2016) in an estuarine area. Since the Columbretes is not subject to direct river influence, such changes must be mainly linked to trawling efforts.

Zooplankton did not follow the “U” pattern described above, and the pteropod *Creseis acicula* experienced a large increase at a depth of 7 cm in MC2, as mentioned above. Due to the pelagic habitat of pteropods, this increase cannot be attributable to trawling. Trends obtained for pteropods at MC2 were consistent with long-term analyses of living pteropods in the NW Mediterranean Sea (1967–2003, Howes et al., 2015). This indicates that pteropods are useful for following climatic variations and chronological reconstructions in sediment cores. In this way, as mentioned, *Creseis* trends also fit with the results from a previous chronological analysis off the Ebro Delta (Cartes et al., 2017). *Creseis* increased in the MC2 profile since the 1970s, linked to higher surface temperatures (0–50-m T in our data) and positive NAO phases. The sharp increase in *C. acicula*, clearly related to surface T, is consistent with the increase in surface water T recorded in recent decades in the western Mediterranean (Vargas-Yáñez et al., 2008, 2009), although with an important reduction in warming rates since the 1990s, also identified in *Creseis* abundance. In addition, high (positive) NAO conditions, also related to *C. acicula*, may lead to better oceanic conditions over the shelf for offshore species, since NAO + is related, in the Mediterranean Sea, to periods of low rainfall (Mariotti et al., 2002) and low advection of POM from coastal waters.

In a preceding study, remains (sclerites) of the bamboo coral *Isidella elongata* showed clear relationships with trawling activities (Cartes et al., 2017). The cessation of trawling on Columbretes Island in 1987 also seems to coincide with an increase in Octocorallia sclerites in the MC2 profile, i.e., at depths of 6–7 cm ca. in 1967–1980, while in older levels beyond the 1970s, the sclerite densities were low. Landings in ports close to the Columbretes Islands (i.e., Castelló) have been available since 1932 and have indicated trawling activity at the shelf. Although trawling probably began earlier off the Castellón coasts, it was during the 1930s when trawling efforts increased significantly in the area, since motorboats began trawling only after 1925. Before ca. 1930s, the fishery efforts off Catalonia (at 70 km to the N of the study area, similar to the dynamics off Castellón) must be considered negligible (Bas et al., 1955). It seems that between 6 and 10 cm (1980–1927), we recorded a decrease in the sclerites of octocorallians, accompanied by a general decrease in benthic molluscs, which may be a consequence of this higher fishing effort. Regarding other factors considered (pollutants), we did not find any clear relationship between the Hg levels and the abundance/diversity of benthos to justify a similar decrease in diversity as found in a semienclosed estuary off Cuba (Armenteros et al., 2016). At the MC2 off the Columbretes, the Hg levels in sediments were close to the background levels reported for Hg in the Mediterranean (30 µg/kg, Angelidis et al., 2011) and were always well below the effective range accepted for marine organisms (150 µg/kg, Long et al., 1995). However, we cannot exclude some chronic impacts of heavy metals, perhaps added to other effects, as suggested by Mauner et al. (2018), since the increase in Hg observed from 10 to 6 cm (likely due to industrialization) coincided with the decrease in benthic molluscs.

The diversity for the whole (benthic) community remained practically constant (approximately 2.5) throughout all MC2 (i.e., over the time period studied). This may be a consequence of the high stability of benthos communities. The species may change, but the diversity remains at a quite constant level. This pattern suggests a well-structured ecosystem with high functional redundancy (de la Torriente et al., 2020). When a species disappears due to a natural or anthropogenic disturbance, another species with the same or even a different functional role can replace it and maintain the levels of diversity and relationships among species. In contrast to the MC4 of the Ebro Delta slope (398 m depth), where mesopelagic fish remains (mainly otoliths) were abundant (Cartes et al., 2017), at MC2, fish, represented mainly by benthic Gobiidae, were too scarce for long-term analysis. Epipelagic fish (small pelagics, sardine or anchovy) were practically absent in MC2, perhaps because they were not very abundant around the Columbretes Islands, as suggested by the low levels of fisheries reported (INDEMARES, 2013).

The eutrophication of Mediterranean waters, which has increased in general since 1960 (Béthoux et al., 2002), may also contribute to the increase in benthos biomass due to higher food availability, especially for filter feeders. Filter feeders were also more linked to low fishing efforts, being more abundant after the cessation of trawling. Such tendencies can also be submitted to more short-term oscillations. The reserve effect, the most important factor affecting changes in the MC2, did not explain why over the last two decades (since ca. 2002), the diversity (H) in the area decreased, both for the whole community and especially for the mobile fauna (fish, crustaceans). Although the Columbretes area is not in an estuary, as was the location in which Armenteros et al. (2016) reported a loss of diversity in bivalves due to river damming, it cannot be excluded that low levels of advection from the mainland, accentuated by the damming of the Ebro River (and other rivers) in the 1950 s-60 s, could also contribute to the decrease in benthos diversity and of (some) mollusc abundances found at the uppermost levels of the MC2. In this way, in the period from 2007 to 2017 (0–2 cm), the Chl *a* concentration (obtained by satellite data consulted) also suffered a drop compared with that of the preceding period (2–3 cm, 2002–2007).

#### 4.3. Conclusion

A detailed analysis of a sediment core from the Columbretes Marine Reserve (MC2, 87 m depth) allowed the identification of a combination of human and climatic variables affecting benthic and pelagic communities in recent decades/centuries. Among the human impacts, the cessation of trawling activities in the 1980 s, after the declaration of an MPA in the region, produced the most evident effect, provoking increases in benthic bivalves and gastropods, changes in the feeding guilds with an increase in filter feeders after the cessation of trawling, and some recolonization by Octocorallia. In contrast, the increase in the pteropod *Creseis acicula* observed since 1995 probably indicates the result of warming in surface waters. These results indicated that the sea is an open system, and within an MPA, changes can be conditioned by climatic and human factors occurring outside of the protected area in addition to factors such as fisheries that can be locally controlled within the MPA. This study based on sediment core reconstruction provides for the first time an historical perspective of the impacts on marine fauna and of the effectiveness of conservation measures after the creation of an MPA.

#### CRedit authorship contribution statement

**J.E. Cartes:** Conceptualization, Validation, Writing – review & editing. **D. Díaz-Viñolas:** Writing – review & editing. **A. Schirone:** Validation, Writing – review & editing. **A. Lombarte:** Validation, Writing – review & editing. **M. Barsanti:** Conceptualization, Validation, Writing – review & editing. **I. Delbono:** Validation, Writing – review & editing. **C. Salas:** Validation, Writing – review & editing. **S. Gofas:**

Validation, Writing – review & editing. **A. Serrano:** Writing – review & editing. **J. Santos-Echeandía:** Validation, 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.

### Data availability

Some data would be potentially available depending of their use and focus to original objectives

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ecolind.2022.109209>.

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