



# Ecological and human health risk assessment of potentially toxic element contamination in waters of a former asbestos mine (Canari, Mediterranean Sea): implications for management

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**Abstract** Between 1948 and 1965, the Canari asbestos mine (Corsica, France) discharged 11 million tonnes of serpentinite rubble into the sea. This study, therefore, aims to assess the environmental and health risks associated with contamination of potentially toxic elements using bioindicators (seagrass and fish) in the areas bordering the former mine within the perimeter of the Cap Corse and Agriate Marine Natural Park. The results and multivariate statistical analyses of the potentially toxic elements, made it possible to identify a concentration gradient, a model of bioaccumulation, and the occurrence of different groups, thus reflecting a spatial variation of the contamination. These results indicate that the former asbestos mine can

still be considered, 55 years after its closure, as a major source of Co, Cr, and Ni for marine ecosystems and still influences the quality of the coastal area today. Our study, therefore, indicates that the two most polluted sites (Albo and Negru) are the closest stations to the south of the old Canary asbestos mine. According to the Trace Elements Pollution Index (TEPI) values, 6 species were classified as having a high contamination level: *Scorpaena notata* (1.37), *Scorpaena porcus* (1.36), *Sepia officinalis* (1.27), *Diplodus vulgaris* (1.02), *Spicara maena* (0.95), and *Mullus surmuletus* (0.94). Regarding the potentially toxic elements measured in the edible tissues of fish, the concentrations were all below the regulatory thresholds and did not reveal any potential risk to human health (Cd, Cu, Fe, Pb, Se, Sn, Zn). This work provides new and useful information to improve the monitoring of the environmental quality of a region characterized by previous mining activity and to assess the potential risk to human health due to the consumption of fish. Beyond the purely scientific aspects, these results could serve as decision support at the regional level for the definition of long-term public policies.

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## Introduction

Natural background levels (NBLs) are defined as concentrations of a given element, species, or chemical

substance present in water controlled by natural geogenic, biological, and atmospheric processes (Griffioen et al., 2008). The natural background level is considered an indicator or a synonym for a geochemical baseline to describe natural variations in component concentrations in the surficial environment (Gao et al., 2020). Several natural factors contribute to the definition of the natural background level, including climate, rainfall composition, frequency, water–gas–rock interactions, and residence time (Sellerino et al., 2019). The definition of NBLs for potentially toxic elements (PTEs) in groundwater and coastal waters from mining environments is a real challenge, as anthropogenic activities boost water–rock interactions, further increasing the naturally high concentrations (Sacchi et al., 2021).

Asbestos minerals can host a large number of potentially toxic elements: nickel, cobalt, chromium, and manganese have been observed at high concentrations in chrysotile asbestos (Isoda et al., 2015). PTEs are natural elements present in the earth's crust, and their multiple industrial, agricultural, pharmaceutical, and technological applications have led to their wide distribution in the environment. Potentially toxic elements released from natural and anthropogenic sources can reach the marine environment, bioaccumulate, and biomagnify throughout the food chain (Atwell et al., 1998). Their use has raised concerns about their potential effects on ecosystems and human health (Tchounwou et al., 2008). Certain PTEs (e.g., selenium, copper, cobalt, molybdenum, manganese, and zinc) are essential to life as they may constitute several key enzymes and play an important role in various redox reactions. However, an excessive amount of these metals can lead to cell and tissue damage (Waalkes et al., 1996). Other PTEs, such as lead, cadmium, mercury, or arsenic, have no established biological function and are considered non-essential and potentially toxic at relatively low concentrations. Some of these contaminants can induce important “cocktail effects” on marine organisms. Unlike some organic pollutants (e.g., polychlorinated biphenyl (PCB), persistent organic pollutants, pesticides, dioxins (POPs)), which can be partially degraded, PTEs are non-degradable and persistent in the environment (Gargouri et al., 2011). Thus, they are considered to be a major source of pollution in the marine environment due to their toxicity, persistence, and ability to accumulate in marine organisms (Bonanno & Di Martino, 2017).

High concentrations of these elements in the different compartments of the ecosystem (e.g., the water column, sediments, and organisms) can lead to profound

ecological consequences such as the loss of living resources, a decrease in biodiversity, and damage to human health (Moore et al., 2004). Monitoring the marine environment with effective tools is therefore a top priority for the implementation of appropriate conservation policies and strategies. Specific species, called bioindicators, are commonly used to monitor the quality of the marine environment and may fulfill specific criteria that make it possible to assess the state of element trace contamination of marine ecosystems (Richir & Gobert, 2016). Among them, *Posidonia oceanica* (L.) Delile is widely recognized as a biomonitor of potentially toxic elements and has been used as a bioindicator for several decades (Richir & Gobert, 2014). Indeed, it presents the characteristics of a good bioindicator: it is sessile, abundant, and easy to collect; it efficiently accumulates pollutants at high levels; it resists pollution; it survives near major sources of contamination; and it reflects the ambient state of its environment (Boudouresque et al., 2012). Thus, their utility as sentinel organisms in PTEs biomonitoring studies is widely recognized. Fish are also recognized as bioindicators of potentially toxic element contamination, providing an integrated overview of the state of their environment (Pleschl et al., 2019). The bioavailability of PTEs in seagrass is associated with several physicochemical parameters, e.g., salinity, temperature, pH, nutrients, organic matter, and light intensity (Bonanno, 2020; Richir et al., 2013). They are regularly used to assess ecological and health risks as they occupy high trophic levels and are an important food source (Agah et al., 2010).

Between 1948 and 1965, the Canari asbestos mine (Corsica, France) discharged 11 million tonnes of serpentinite mining waste into the sea (Bernier et al., 1997). In addition, high concentrations of PTEs are also present in the region in the rocks hosting the chrysotile. Many studies have identified high concentrations of PTEs in surface and groundwater from serpentinite, so it is likely that pollution in the area is related to both the discharge of crushed serpentinite rock and asbestos. More than 50 years after its closure, contaminations by cobalt (Co), chromium (Cr), and nickel (Ni) were detected nearby within different biological compartments (Lafabrie et al., 2007). These elements can be considered “tracers” of mining activity. Several previous studies have already studied the impact of this contamination on the marine environment (Andral et al., 2004; Gosselin et al., 2006;

Lafabrie et al., 2009; Ternengo et al., 2018). This study, therefore, aims to assess the environmental and health risks associated with the contamination of PTEs in the areas bordering the former mine within the perimeter of the Cap Corse and Agriate Marine Natural Park (PNMCCA). More precisely, the objectives of this study are to: (i) study the spatial and temporal variations of the concentrations of 16 PTEs, with particular attention to the tracers of mining (e.g., Co, Cr, Ni), (ii) assess the quality of the coastal area through different bioindicators (seagrass, fish), characterizing the area of influence of the former mine (contamination gradient), (iii) determine whether these PTEs persist within the marine ecosystem more than 50 years after the closure of the mine, identifying whether there are phenomena of bioaccumulation/bioreduction, and (iv) estimate the quality of the flesh from commercial species caught near the Canari site, quantifying the potential risks to human health linked to their consumption by the local population.

## Materials and methods

### Study area

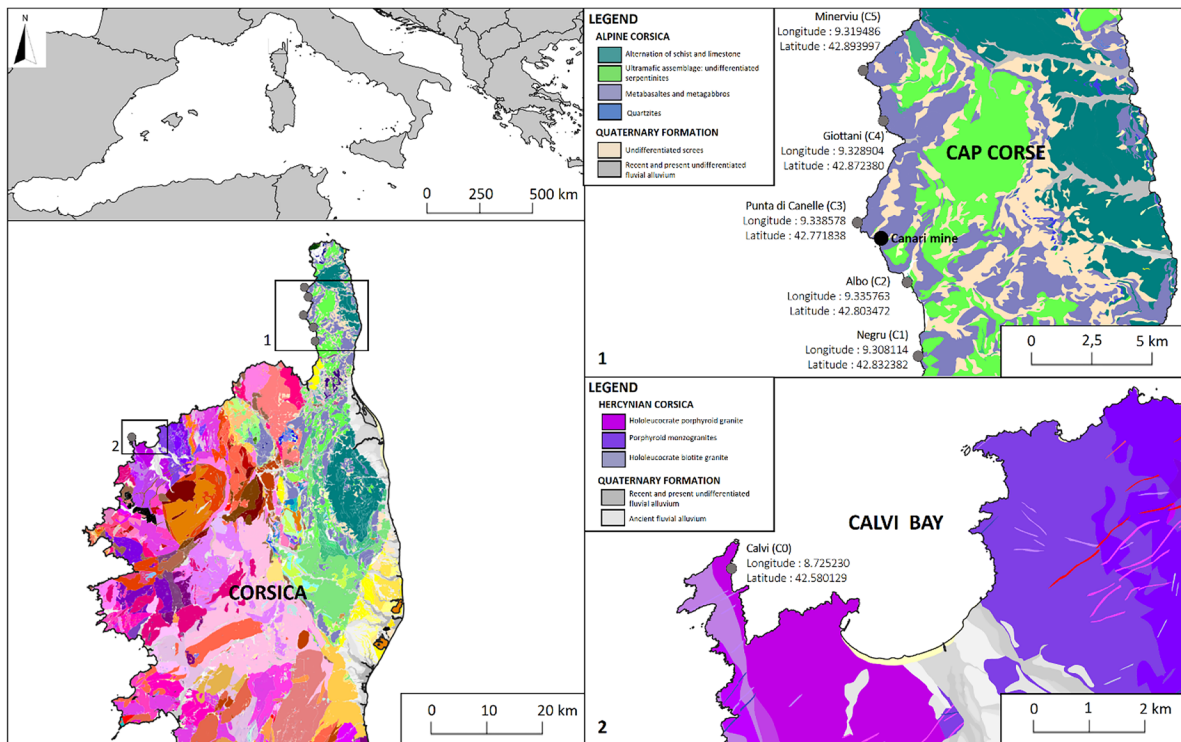
The present study was performed around the island of Corsica (France; NW Mediterranean), which is located southeast of the French mainland and north of Sardinia (Italy) (42°N and 9°E). Corsica is the fourth largest island in the Mediterranean Sea and is characterized by a mountainous landscape and a disparate underwater morphology, featuring a steep descent (down to 3000 m depth, 10 km offshore) along the western part of the island (Pluquet, 2006) and wide expanses of relatively shallow waters (depths of <200 m) along the east coast. Corsica has a surface area of 8722 km<sup>2</sup> and a coastline of 1047 km, representing more than half of the French Mediterranean coast. Corsica is divided into two distinct geological regions:

- Hercynian Corsica, which corresponds to a Paleozoic basement largely intruded by calc-alkaline granitoids of Carboniferous age and Permian alkaline granites (Faure et al., 2015; Li et al., 2014). This base is surmounted by an Eocene sedimentary cover. Covering about three-quarters of the area of the island, Hercynian Corsica corresponds to the eastern and southern parts (Magott, 2016).

- Alpine Corsica corresponds to Cap Corse and the northeast part of the island (Magott, 2016). It is mainly made up of ophiolitic rocks, sometimes associated with their sedimentary cover, and allochthonous carbonates of continental origin that underwent HP/BT-type metamorphism of variable intensity during the Alpine orogeny (Lahondère, 1991). The ophiolitic formations of Alpine Corsica are full of asbestos and can locally present a health risk for the populations concerned.

### Sampling procedures

*P. oceanica* is a protected and endemic species of the Mediterranean Sea that provides a variety of ecosystem services. It is the most common bio-indicator species on the study site. For the *Posidonia oceanica* sampling, 6 sites were selected along the north-western coast of Corsica in order to account for the heterogeneous oceanographic and geological properties (current, sediment drift, nature of the seabed, depth, distance): Minerviu (8.6 km north of the mine, C5), Giottani (5.5 km north of the mine, C4), Punta di Canelle (1.8 km north of the mine, C3), Albo (2.3 km south of the mine, C2), Negru (6.6 km south of the mine, C1), and Calvi (55 km south of the mine, C0) (Fig. 1). Such a sampling strategy was put in place in order to obtain as much information as possible on the dispersion or potential dilution of pollutants along a North–South contamination gradient. Sampling was carried out during the summer of 2019. On each of the stations, 20 *P. oceanica* shoots ( $n=120$ ) were collected by scuba divers at a depth of 15 m using the non-destructive method (Non-Destructive Shoot Sampling Method (NDSM)) (Gobert et al., 2020). This technique involves cutting all of the leaves of a *P. oceanica* shoot (with scissors) just above the scale visible outside the shoot. NDSM prevents damaging the leaf meristem and ensures the post-sampling regrowth of cut leaves. At the STARESO laboratory (Corsica, France), the epiphytes were removed from the *P. oceanica* leaves using a ceramic scalpel to prevent metallic contamination. Only adult leaves were taken into account because it has been reported in the literature that potentially toxic elements preferentially accumulate in them (Lafabrie et al., 2007). The adult leaves (between 2 and 5 leaves per shoot) were grouped together by the site to constitute pools in order to have sufficient biological material for the analysis. The samples were rinsed with



**Fig. 1** A geological map of the area showing the location of the collection sites for *Posidonia oceanica* meadow samples

water, frozen ( $-20\text{ }^{\circ}\text{C}$ ), oven-dried ( $60\text{ }^{\circ}\text{C}$  for 48 h), and then reduced manually to a powder in an agate mortar.

The fish samples were collected during the summer of 2019 (August, September) in the immediate vicinity of the site of the former Canari mine during two experimental fisheries using trammel nets in collaboration with an artisanal fisherman from Cap Corse (Fig. 2). The objective is to target and select sedentary sentinel species that are regularly consumed by the local population. In respect of animal welfare and ethical rules, the fish were slaughtered by immersion in ice water (hypothermia). In total, 144 individuals were collected, representing fish (and mollusk) from 23 different species, as detailed in Table 1. Each fish was measured (total length (LT)). The specimens were dissected in the laboratory using ceramic scissors to avoid contamination. The scales were removed, and the flesh was cut into small pieces to homogenize the sample. From each individual, a piece of muscle, the most commonly consumed part of a fish, weighing an average of 10 g (wet weight), was collected. Each tissue sample was immediately stored in individual

plastic bags and frozen at  $-25\text{ }^{\circ}\text{C}$ . The samples were then lyophilized and reweighed (dry weight). The lyophilized tissues were crushed and stored in individual plastic vials until elemental analysis.

#### Potentially toxic element analysis

To analyze the potentially toxic elements, the samples were first mineralized. A total of 180 mg of each sample was placed in Teflon-coated digestion bombs. Reagents added for digestion included 40% (1 ml) of hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) and 60% (2 ml) of nitric acid ( $\text{HNO}_3$ ). After digestion in a laboratory microwave (Ethos D, Milestone Inc.), the samples were diluted in ultra-pure water (50 ml of Falcon). The analyses of the 16 potentially toxic elements (Ag, As, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb, Se, Sn, V, and Zn) were carried out by inductively coupled plasma mass spectrometry (ICP-MS) using the dynamic reaction cell technology (ELAN DRC II, Perkin Elmer®). These analyses were carried out at the Oceanology Laboratory of the University of Liège according to the method described in Richir and



**Fig. 2** Illustrative picture showing the former asbestos mine of Canari (Corsica, France)



Gobert (2014). To verify that the chemicals used during the treatment of the samples could not be the source of contamination in PTEs, blanks of these chemicals were analyzed. Analytical quality control was performed using DORM4- (fish protein) and NIST2976-certified (mussel tissue) reference materials for fish and BCR60 (*Lagarosiphon major*), BCR61 (*Platihyphnidium riparinoides*), and BCR62 (*Olea europaea*) for *P. oceanica*. These analyses revealed a high level of agreement between the certified values for all potentially toxic

elements (the overall mean recovery was  $94 \pm 12\%$ ). The limits of detection and quantification were calculated for each potentially toxic element on the basis of the distribution of the measurements on their respective blank (Currie, 1999). Concentrations that were found to be below their analytical detection limit were considered half of the detection limit value during the statistical processing of the data. Results were expressed as milligrams per kilogram of wet weight for fish and milligrams per kilogram of dry weight for *P. oceanica*.

**Table 1** Characteristics of the 23 species of fish sampled near the former asbestos mine in Canari ( $N$  total = 144)

Common name	Scientific name	$N$	Length (cm)
Red scorpionfish	<i>Scorpaena scrofa</i>	1	19
Atlantic horse mackerel	<i>Trachurus trachurus</i>	1	28
Brown meager	<i>Sciaena umbra</i>	3	24
East Atlantic peacock wrasse	<i>Symphodus tinca</i>	18	21
Black seabream	<i>Spondyliosoma cantharus</i>	5	23
Greater weever	<i>Trachinus draco</i>	4	24
Streaked gurnard	<i>Chelidonichthys lastoviza</i>	3	27
Brown wrasse	<i>Labrus merula</i>	4	26
Green wrasse	<i>Labrus viridis</i>	1	26
Blotched picarel	<i>Spicara maena</i>	15	20
Forkbeard	<i>Phycis phycis</i>	4	34
Common pandora	<i>Pagellus erythrinus</i>	4	22
Small red scorpionfish	<i>Scorpaena notata</i>	7	12
Black scorpionfish	<i>Scorpaena porcus</i>	8	16
Wide-eyed flounder	<i>Bothus podas</i>	1	13
Striped red mullet	<i>Mullus surmuletus</i>	13	21
Common two-banded sea bream	<i>Diplodus vulgaris</i>	31	17
Common cuttlefish	<i>Sepia officinalis</i>	1	19
Comber	<i>Serranus cabrilla</i>	2	12
Painted comber	<i>Serranus scriba</i>	8	19
Annular seabream	<i>Diplodus annularis</i>	2	16
Stargazer	<i>Uranoscopus scaber</i>	5	19
Starry weever	<i>Trachinus radiatus</i>	3	23
<b>Total</b>		144	

$N$ , number of individuals sampled

## Pollution indices for ecological risks

### Trace Element Pollution Index

The level of contamination for each station was calculated using the “Trace Element Pollution Index” (TEPI) developed by Richir and Gobert (2014). The TEPI is a monitoring tool for evaluating the overall state of the chemical contamination of the coastal area. The TEPI makes it possible to differentiate and characterize little or highly contaminated sites. It also allows the reliable comparison of the levels of overall contamination in PTEs between environmental studies, regardless of the list of contaminants assayed and/or the bioindicator species used. Once the TEPI values have been calculated, they can be classified qualitatively according to a 3-level contamination scale, defined according to the quartile method: (1) low contamination level (TEPI values lower than the average of the 1st quartile), (2)

medium contamination level (TEPI values between the average of the 1st and 3rd quartile), and (3) high contamination level (TEPI values higher than the average of the 3rd quartile) (Morrison et al., 2017). The higher the TEPI value, the more polluted a site is.

$$\text{TEPI} = (\text{Cf}_1 * \text{Cf}_2 \dots \text{Cf}_n)^{1/n}$$

where  $\text{Cf}_n$  is the normalized mean concentration of the PTE at a given site and  $n$  is the total number of examined potentially toxic elements.

### Trace Element Spatial Variation Index

The “Trace Element Spatial Variation Index” (TESVI) gives an overall view of the spatial variability for each TE between stations (Richir & Gobert, 2014). For a given PTE, the higher the value of the TESVI, the more its environmental levels will vary overall in the

study area. In order to compare and order the PTEs according to the spatial variability of their environmental levels, the TESVI was calculated as follows:

$$TESVI = \left[ \left( \frac{x_{\max}}{x_{\min}} \right) \left( \frac{\sum(x_{\max}/x_i)}{n} \right) \right] * SD$$

where  $x_{\max}$  and  $x_{\min}$  are the maximum and minimum average concentrations recorded among the  $n$  stations,  $x_i$  is the average concentrations recorded in each of the  $n$  stations, and  $SD$  is the standard deviation of the average ratio  $\sum(x_{\max}/x_i)/n$ .

Pollution indices for human health risks

*The admissible regulatory thresholds*

To assess the potential risks to human health associated with the consumption of seafood, several international health safety institutions (e.g., WHO, FAO, JECFA, USEPA) have established permissible regulatory thresholds/maximum limits not to be exceeded. These levels are expressed in milligrams per kilogram of fresh weight and are 95 (Ba), 0.3 (Cd), 1 (Cr), 20 (Cu), 10 (Fe), 2 (Fe), 0.3 (Pb), 2 (Se), 250 (Sn), and 50 (Zn) (FAO/WHO, 2013, 2014; JECFA, 2011; USEPA, 2010).

*Estimated weekly intake/provisional tolerable weekly intake*

The estimated weekly intake estimates the amount per unit of body weight of a contaminant in food that can be ingested over a lifetime without risk to health. The concept of estimated weekly intake (EWI) takes into account daily variations in patterns of human consumption. It provides a basis on which the concentrations obtained can be compared with the provisional tolerable weekly intake (PTWI) defined by the Joint FAO/WHO Expert Committee. An EWI lower than the PTWI means that there is no significant health risk for the exposed population. The risk of consumption of potentially toxic elements (weekly) is estimated by calculating the respective levels observed in fish for a person weighing 70 kg and a consumption rate of 427 g/week (defined for the European population). Thus, the EWI can be determined using the following equation:

$$\text{Estimated weekly intake: } EWI = (C_m * IR_w) / BW$$

where  $C_m$  represents the concentration of potentially toxic elements in fish ( $\mu\text{g}/\text{kg}^{-1}$ ),  $IR_w$  is the weekly ingestion rate (kg), and  $BW$  is the body weight (kg).

*Target hazard quotient, total target hazard quotient*

The target hazard quotient (THQ) is used to assess the risk of developing so-called non-carcinogenic diseases due to the consumption of seafood contaminated with potentially toxic elements. THQ incorporates not only potentially toxic element intake, but also other important data, such as frequency and duration of exposure, rate of ingestion, and average body weight of the population. THQ is the ratio of exposure to reference doses. A ratio less than 1 indicates no obvious risk and that the level of daily exposure is unlikely to cause adverse effects in a person's lifetime. If the exposure level exceeds this threshold, potential non-carcinogenic effects may be of concern in the contaminated population (Xiao et al., 2021). High THQ values mean a higher likelihood of experiencing long-term non-carcinogenic effects.

The THQ was calculated according to the methodology of the United States Environmental Protection Agency (USEPA, 2010):

$$THQ = \frac{EF * ED * IR * C}{RfD * BW_a * AT_n} * 10^{-3}$$

where  $EF$  and  $ED$  represent respectively the frequency of exposure (365 days/year) and the average lifespan (70 years);  $IR$  is the rate of fish consumption (60 g/day for one person; FAO (2005));  $C$  is the concentration of potentially toxic elements ( $\text{mg kg}^{-1}$ );  $RfD$  is the oral reference dose in milligrams per kilogram. The oral reference doses for Zn, Cu, Pb, Cd, Fe, Cr, Mn, and Se were estimated to be 0.3, 0.04, 0.00357, 0.001, 0.7, 0.003, 0.14, and 0.005, respectively.

Since exposure to multiple pollutants can lead to combined or synergistic effects, the total target hazard quotient (TTHQ) was also calculated as the arithmetic sum of each THQ value:

$$TTHQ = THQ(\text{PTE } 1) + THQ(\text{PTE } 2) + THQ(\text{PTE } n)$$

*Carcinogenic risk assessment*

The carcinogenic risk assessment is used to estimate the potential risk of developing cancer from exposure



to pollutants such as Pb (Islam et al., 2015). Carcinogenic risks are estimated as the probability that an individual develops cancer during their lifetime following exposure to a potential risk factor. Risk levels range from  $10^{-4}$  (indicating a 1 in 10,000 chance that an individual will develop cancer) to  $10^{-6}$  (indicating a 1 in 1,000,000,000 chance that an individual will develop cancer). There are four categories of carcinogenic risk (CR)  $\leq 10^{-6}$  = low;  $10^{-4}$  to  $10^{-3}$  = moderate;  $10^{-3}$  to  $10^{-1}$  = high; and  $\geq 10^{-1}$  = very high.

$$CR = (EF \times ED \times IR \times C \times CPFo) / (BW \times AT)$$

where EF is the frequency of exposure (365 days/year); ED is the duration of exposure (70 years, average lifespan); IR is the rate of food ingestion; *C* is the concentration of a potentially toxic element in fish; BW is the average body weight of an adult (70 kg); and AT is the average exposure time for non-carcinogenic agents (365 days / year  $\times$  ED, assuming 70 years). CPFo is the oral carcinogenic slope factor (USEPA, 2010).

### Statistical analyses

Normality and homogeneity of variances were tested using the Kolmogorov–Smirnov and Levene tests, respectively. To better meet the assumptions of standard parametric statistical tests, to reduce the effect of outliers biasing the distribution of the data, and to bring the elemental concentrations back to the same range, the data were transformed into a natural logarithm. Statistical differences in potentially toxic element concentrations between sampling sites were examined using parametric ANOVA analysis of variance and Tukey post hoc tests. A Pearson correlation analysis was conducted to identify possible correlations between potentially toxic elements (inter-metal matrix), fish lengths, and trophic level values of different species. A principal component analysis (PCA) was performed for the study of observations in a two-dimensional space in order to identify homogeneous groups of observations and to visualize the correlations between the variables. In detail, a PCA was used to obtain detailed information about the dataset and gain insight into the distribution of potentially toxic elements by detecting similarities or differences in samples. Further classification was performed via hierarchical ascendant classification (HAC, cluster analysis), using centroid clustering methods and Euclidean distance for the measure of dissimilarity. The principle of the HAC is to bring together individuals

according to a criterion of resemblance defined beforehand, which will be expressed in the form of a matrix of distances, expressing the distance existing between each individual taken two by two. Two identical observations will have a zero distance. The more dissimilar the two observations, the greater the distance. This classification method makes it possible to visualize the progressive grouping of data using a dendrogram. For all statistical tests, the results are considered significant if a level of significance of  $p < 0.05$  is reached.

## Results

### Ecological risks

The results of the mean concentrations of PTEs in the leaves of *P. oceanica* are presented in Table 2. The distribution pattern of the average potentially toxic element concentrations generally followed the decreasing order: Fe > Zn > Mn > Ni > Cu > Co > V > Cd > Mo > Pb > Cr > As > Se > Ag > Sb > Sn. There is a very large variation in the concentrations between sampling stations. Significant differences between stations were found for a large number of PTEs (Table 2). Out of the 16 PTEs analyzed, 2 of them (Ag and Cu) did not show significant differences between the 6 sites (ANOVA test,  $p > 0.05$ ), while the 14 others showed significant differences among sites (ANOVA test,  $p < 0.05$ ) (Table 2). The highest concentrations of cobalt (Co), chromium (Cr), and nickel (Ni) were observed in samples from the Albo site (C2) located in the immediate vicinity of the former asbestos mine ( $9.68 \pm 1.49$  mg kg<sup>-1</sup> of dry weight;  $2.90 \pm 0.90$  mg kg<sup>-1</sup> of dry weight;  $75.43 \pm 12.05$  mg kg<sup>-1</sup> of dry weight for Co, Cr, and Ni, respectively). These values were significantly higher than those measured at the 5 other sites ( $p < 0.001$ ). On the contrary, the minimum values for these potentially toxic elements were found mainly on the site of Calvi ( $1.95 \pm 0.31$ ,  $0.39 \pm 0.08$ , and  $26.16 \pm 5.29$  mg kg<sup>-1</sup> of dry weight for Co, Cr, and Ni, respectively) (Table 2).

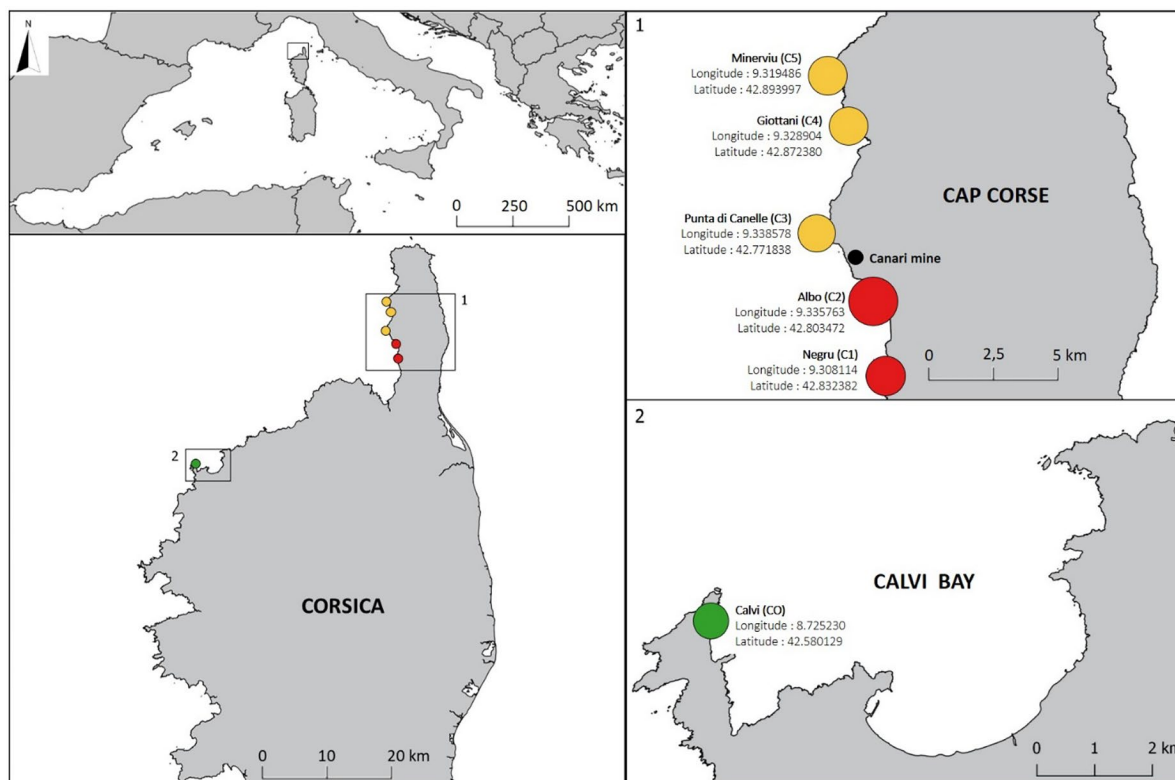
For each station, the TEPI was calculated and used to compare the total content of PTEs at the different sampling sites. TEPI values ranged from 1.29 at Albo (C2) to 0.79 at Calvi (C0). Three stations were classified with an average level of contamination: Minerviu, Giottani, and Canelle (TEPI: 0.89, 0.86, and 0.87, respectively) (Fig. 3; Table 3). According to TEPI, Albo and Negru were the



**Table 2** Average concentration of potentially toxic elements (expressed in mg/kg per dry weight,  $\pm$  standard deviation calculated on the samples of the same site) in the adult leaves of *P. oceanica* in the study area (Calvi-Cap Corse)

Element	Calvi (C0)	Negru (C1)	Albo (C2)	Canelle (C3)	Giottani (C4)	Minerviu (C5)
Ag	0.39 ( $\pm$ 0.19) a	0.33 ( $\pm$ 0.10) a	0.38 ( $\pm$ 0.09) a	0.52 ( $\pm$ 0.29) a	0.48 ( $\pm$ 0.20) a	0.43 ( $\pm$ 0.18) a
As	0.80 ( $\pm$ 0.11) a	0.57 ( $\pm$ 0.38) bc	0.83 ( $\pm$ 0.30) a	0.67 ( $\pm$ 0.15) ab	0.70 ( $\pm$ 0.09) a	0.43 ( $\pm$ 0.15) c
Cd	1.65 ( $\pm$ 0.31) a	1.60 ( $\pm$ 0.29) a	1.13 ( $\pm$ 0.24) b	2.47 ( $\pm$ 0.64) c	2.12 ( $\pm$ 0.39) c	2.32 ( $\pm$ 0.59) c
Co	1.95 ( $\pm$ 0.31) a	4.87 ( $\pm$ 0.96) b	9.68 ( $\pm$ 1.49) c	3.80 ( $\pm$ 0.89) d	3.55 ( $\pm$ 0.52) d	5.10 ( $\pm$ 1.08) b
Cr	0.39 ( $\pm$ 0.08) a	0.97 ( $\pm$ 0.42) b	2.90 ( $\pm$ 0.90) c	0.36 ( $\pm$ 0.08) a	0.41 ( $\pm$ 0.09) a	0.86 ( $\pm$ 0.24) b
Cu	5.98 ( $\pm$ 2.18) a	5.75 ( $\pm$ 1.65) a	6.37 ( $\pm$ 1.37) a	8.10 ( $\pm$ 4.36) a	8.27 ( $\pm$ 5.03) a	7.18 ( $\pm$ 2.84) a
Fe	51.88 ( $\pm$ 11.72) a	75.90 ( $\pm$ 23.65) b	116.80 ( $\pm$ 26.07) c	38.81 ( $\pm$ 8.71) d	47.95 ( $\pm$ 10.75) ad	56.97 ( $\pm$ 12.64) a
Mn	39.77 ( $\pm$ 10.55) a	43.16 ( $\pm$ 11.26) ab	65.76 ( $\pm$ 10.59) c	50.13 ( $\pm$ 9.54) bd	49.98 ( $\pm$ 8.42) bd	57.36 ( $\pm$ 12.05) cd
Mo	1.50 ( $\pm$ 0.39) a	1.98 ( $\pm$ 0.53) bc	2.45 ( $\pm$ 0.44) c	1.66 ( $\pm$ 0.55) ab	1.98 ( $\pm$ 1.22) ab	1.61 ( $\pm$ 0.32) ab
Ni	26.16 ( $\pm$ 5.29) a	37.99 ( $\pm$ 7.95) bc	75.43 ( $\pm$ 12.05) d	31.44 ( $\pm$ 7.96) ae	32.20 ( $\pm$ 5.83) be	45.60 ( $\pm$ 10.08) c
Pb	1.56 ( $\pm$ 0.33) ab	2.66 ( $\pm$ 2.01) b	1.31 ( $\pm$ 0.52) a	1.96 ( $\pm$ 1.60) ab	1.82 ( $\pm$ 1.43) ab	1.49 ( $\pm$ 0.58) a
Sb	0.26 ( $\pm$ 0.05) a	0.31 ( $\pm$ 0.05) a	0.43 ( $\pm$ 0.08) b	0.27 ( $\pm$ 0.06) a	0.29 ( $\pm$ 0.07) a	0.29 ( $\pm$ 0.07) a
Se	0.45 ( $\pm$ 0.08) a	0.47 ( $\pm$ 0.10) a	0.63 ( $\pm$ 0.11) b	0.41 ( $\pm$ 0.10) ac	0.37 ( $\pm$ 0.05) c	0.47 ( $\pm$ 0.08) b
Sn	0.03 ( $\pm$ 0.01) ab	0.03 ( $\pm$ 0.01) b	0.02 ( $\pm$ 0.01) ac	0.02 ( $\pm$ 0.01) ac	0.02 ( $\pm$ 0.01) cd	0.02 ( $\pm$ 0.01) d
V	1.79 ( $\pm$ 0.71) a	1.80 ( $\pm$ 1.72) a	5.39 ( $\pm$ 5.35) b	0.99 ( $\pm$ 1.03) c	0.71 ( $\pm$ 0.33) c	0.84 ( $\pm$ 0.33) c
Zn	48.09 ( $\pm$ 7.68) a	52.31 ( $\pm$ 11.36) ab	48.02 ( $\pm$ 11.23) a	67.47 ( $\pm$ 16.70) c	59.31 ( $\pm$ 10.16) bc	55.75 ( $\pm$ 14.12) abc

Different letters (abcd) denote significant differences between groups (ANOVA + Tukey HSD test,  $p < 0.05$ )



**Fig. 3** Map of the study area and cartographic representation of the Trace Element Pollution Index (TEPI) concerning *Posidonia oceanica* samples on the 6 sites studied (Calvi-Cap

Corse area). Color scale: red, high contamination level; yellow, medium contamination level; green, low contamination level

most contaminated sites with the highest levels of Co, Cr, and Ni. The minimum TEPI value was recorded for the Calvi site, with a low level of contamination of 0.79. This allows us to establish the order from the most contaminated site to the least contaminated site as follows: Albo > Negru > Minerviu > Giottani > Canelle > Calvi.

The resulting TESVI values ranged from 0.19 to 8.12 (Table 4). Cr is the element that showed the greatest spatial variation between sites, with the highest value of TESVI (8.12). The elements that have high

variability between sites are Cr, V, Co, and Fe with TESVI values ranging from 8.12 to 1.31. Zn, Cu, Ag, and Mo are the elements that show the smallest spatial variation between sites, with TESVI values ranging from 0.19 to 0.30. The other elements As, Cd, Mn, Ni, Pb, Sb, Se, and Sn have an intermediate level of spatial variation (TESVI between 0.30 and 1.24). The TESVI values have been listed in decreasing order: Cr, V, P, Co, Fe, Ni, Cd, Sn, Pb, As, Sb, Se, Mn, Mo, Ag, Cu, and Zn (Table 4). It appears that out of the 16 elements

**Table 3** Value of the Trace Element Pollution Index (TEPI) for the 6 stations sampled in the Calvi-Cap Corse area concerning the *P. oceanica* meadow

Site	TEPI	Quality scale	Color scale
Minerviu (C5)	0.89	Medium contamination level	Yellow
Giottani (C4)	0.86	Medium contamination level	Yellow
Canelle (C3)	0.87	Medium contamination level	Yellow
Albo (C2)	1.29	High contamination level	Red
Negru (C1)	0.99	High contamination level	Red
Calvi (C0)	0.79	Low contamination level	Green

**Table 4** Trace Element Spatial Variation Index (TESVI) of the 16 potentially toxic elements analyzed in the adult leaves of *Posidonia oceanica* at 6 sites in the Calvi-Cap Corse area

Element	$x_{max}/x_{min}$	$\sum(x_{max}/x_i)/n$	TESVI	Station	Color scale
Cr	8.15	0.98	8.12	Albo	
V	7.64	1.91	7.04	Albo	
Co	4.97	4.82	2.71	Albo	
Fe	3.00	64.71	1.31	Albo	
Ni	2.88	41.47	1.24	Albo	
Cd	2.18	1.88	0.58	Canelle	
Sn	2.10	0.02	0.54	Negru	
Pb	2.02	1.79	0.54	Negru	
As	1.91	0.66	0.42	Albo	
Sb	1.63	0.30	0.33	Albo	
Se	1.71	0.46	0.33	Albo	
Mn	1.65	51.02	0.30	Albo	
Mo	1.63	1.86	0.30	Albo	
Ag	1.55	0.42	0.25	Canelle	
Cu	1.43	6.93	0.22	Canelle	
Zn	1.40	55.15	0.19	Canelle	

The higher the value of TESVI, the greater the spatial variation of this element between study sites: red, high level of variation; yellow, intermediate level of variation; green, low level of variation

analyzed, the Albo site was identified 10 times as the station where the maximum concentrations of these elements were observed (As, Co, Cr, Fe, Mn, Mo, Ni, Sb, Se, V) (Table 4). The maximum concentrations of Ag, Cd, Cu, and Zn were recorded at the Canelle site, and those of Pb and Sn at the Negru site (Table 4).

PCA was used to discriminate the different stations according to the concentrations of PTEs in the leaves of *P. oceanica*. The first two axes of the PCA cumulatively explain 59.2% of the total variance (Fig. 4). Samples from the Albo site are grouped in the upper right quadrant and are clearly separated from other sites. The Albo site appears to correlate with the concentrations of V, Se, Cr, Fe, Co, and Ni. For the other 5 sites, no clear differentiation emerged from the PCA. Nevertheless, the sites of Canelle, Giottani, and Minerviu seem to be linked to high concentrations of Pb, Cd, Ag, Cu, and Zn (Fig. 4).

HAC analysis, based on the potentially toxic element concentration distributions by site, identified three distinct groups (clusters I, II, and III) (Fig. 5). Cluster I is associated primarily with samples from Albo and Negru, the closest sites south of the former asbestos mine. These sites are characterized by the

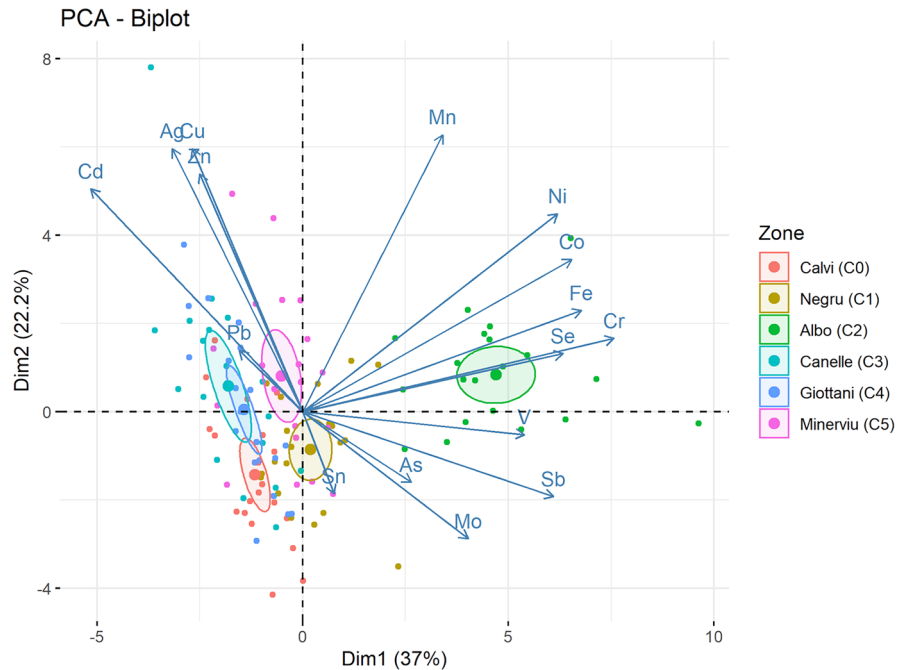
highest concentrations of contaminants. In detail, cluster I consists of 100% of Albo samples and 20% of Negru samples. Cluster II mainly brings together samples from the Calvi site (65%) with a mixture of samples from Negru, Canelle, Giottani, and Minervia. Finally, cluster III is mainly associated with the Minerviu (85%), Canelle (65%), Negru (65%), and Giottani (60%) sites, located to the north and at an average distance from the former asbestos mine.

Human health risks

Table 5 presents the average concentrations of the 16 PTEs analyzed in the muscle of the 23 species of fish and mollusks. The distribution diagram of the average concentrations follows the following decreasing order: Zn > Fe > As > Se > Mn > Cu > Pb > Ni > V > Cr > Co > Mo > Cd > Sn > Sb > Ag. The results of ANOVA and Tukey’s multiple post hoc comparison tests showed significant differences ( $p < 0.05$ ) in PTE concentrations among species.

For each species, the TEPI was calculated and was used to compare the total TE content considering all individuals (Table 6). The TEPI values ranged from a maximum of 1.37 in *Scorpaena notata* to a minimum

**Fig. 4** Two-dimensional representation of the principal component scores for *Posidonia oceanica* sample sites (Calvi-Cap Corse area) differentiated according to potentially toxic element concentrations



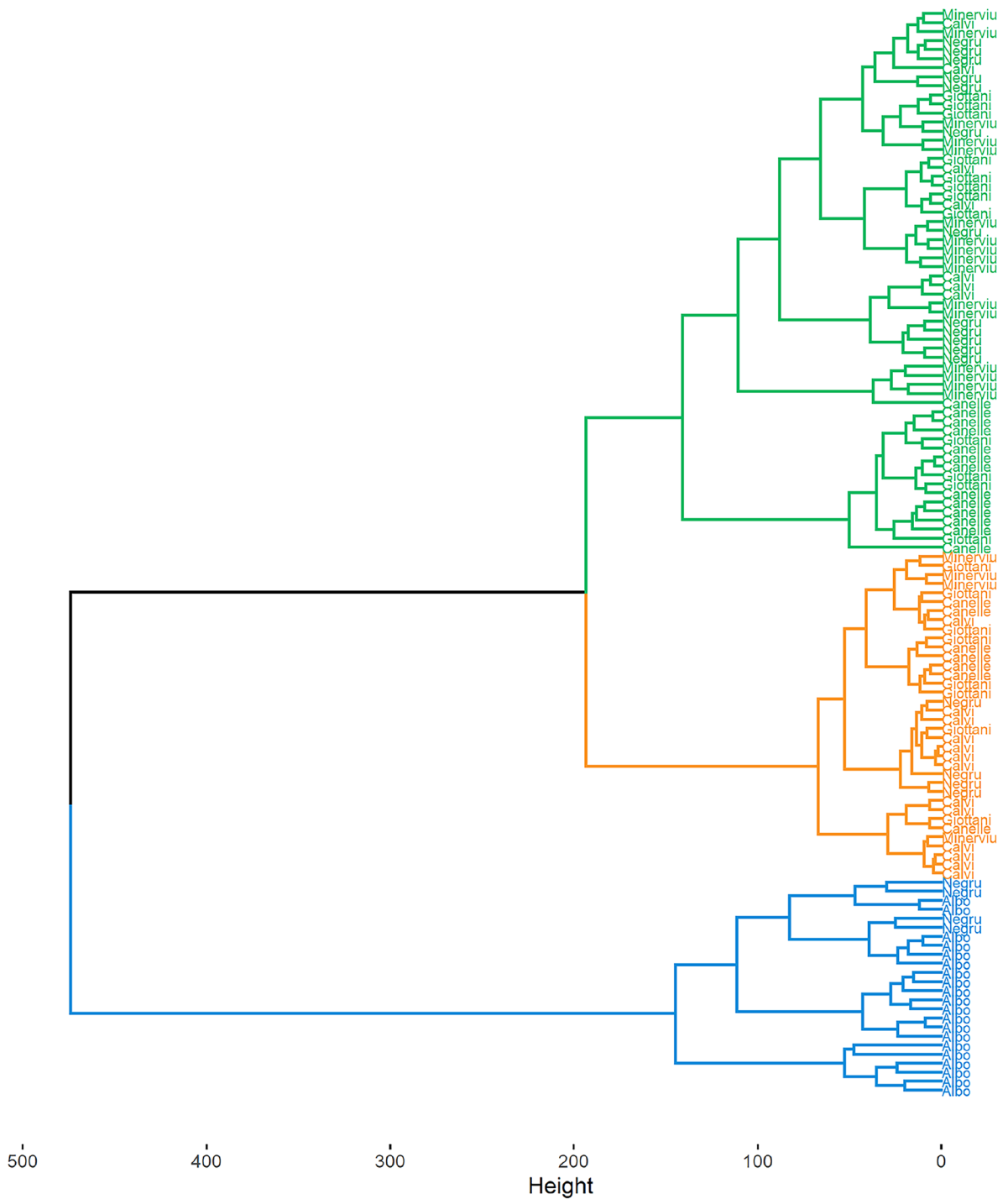
of 0.39 in *Trachinus draco*. According to the TEPI values, 6 species were classified as having a high contamination level: *Scorpaena notata* (1.37), *Scorpaena porcus* (1.36), *Sepia officinalis* (1.27), *Diplodus vulgaris* (1.02), *Spicara maena* (0.95), and *Mullus surmuletus* (0.94). Eleven species were classified as having a medium contamination level: *Symphodus tinca* (0.88), *Serranus cabrilla* (0.87), *Diplodus annularis* (0.85), *Serranus scriba* (0.82), *Sciaena umbra* (0.63), *Labrus merula* (0.57), *Scorpaena scrofa* (0.56), *Trachurus trachurus* (0.56), *Bothus podas* (0.54), *Trachinus radiatus* (0.52), and *Uranoscopus scaber* (0.51). The minimum TEPI values, which represent a low contamination level, were recorded for 6 species: *Chelidonichthys lastoviza* (0.50), *Spondylisoma cantharus* (0.46), *Labrus viridis* (0.45), *Phycis phycis* (0.42), *Pagellus erythrinus* (0.40), and *Trachinus draco* (0.39). This allows us to establish the order of the most contaminated species to the least contaminated species as follows: *Scorpaena notata*, *Scorpaena porcus*, *Sepia officinalis*, *Diplodus vulgaris*, *Spicara maena*, *Mullus surmuletus*, *Symphodus tinca*, *Serranus cabrilla*, *Diplodus annularis*, *Serranus scriba*, *Sciaena umbra*, *Labrus merula*, *Scorpaena scrofa*, *Trachurus trachurus*, *Bothus podas*, *Trachinus radiatus*, *Uranoscopus scaber*, *Chelidonichthys lastoviza*, *Spondylisoma cantharus*, *Labrus viridis*, *Phycis phycis*, *Pagellus erythrinus*, and *Trachinus draco* (Table 6).

The relationship between fish length and mean PTE concentrations and inter-elemental correlations were investigated (Fig. 6). Significant negative correlations were found between the concentrations of most PTEs (12/16 studied) and the size of the fish (Fig. 6). Thus, for the vast majority of PTEs, their concentrations decreased with increasing body size. As regards the inter-elementary relationships, strong and significant positive correlations were found, for example, between Co and Ni, As and Co, or even Pb and Cd (Fig. 6). Significant negative correlations were also found between Zn and As, Se and Cu, and Pb and Co (Fig. 6).

Maximum safety level set for a given element by international agencies were used to assess the human risk associated with the consumption of fish. These levels were respectively 0.3, 1, 20, 10.2, 0.5, 0.3, 2, 250, and 50 mg kg<sup>-1</sup> for Cd, Cr, Cu, Fe, Ni, Pb, Se, Sn, and Zn (European Commission, 2006; JECFA, 2011; MAFF, 2000; FAO/WHO, 2014). In our study, the average levels were all below the standard safety values for fish intended for human consumption.

The EWI values are compared to the recommended PTWI values (Table 7). The EWI values range from 0.01 for Pb to 47.1 for Zn. The EWI of Cu, Cr, Cd, Pb, and Zn for a 70-kg adult consuming 470 g of fish per week throughout the year were all below the limits set





**Fig. 5** Hierarchical ascendant classification based on the potentially toxic element concentration at each site (Calvi-Cap Corse area) in *Posidonia oceanica* meadows

**Table 5** Average concentrations of potentially toxic elements (expressed in mg/kg per wet weight) in the muscles of fish sampled in the study area (Cap Corse)

Species	Ag	As	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	Sb	Se	Sn	V	Zn
<i>Scorpaena scrofa</i>	0.0007 ab	1.5657 abc	0.0020 ab	0.0001 a	0.0455 a	0.1809 ab	2.9368 a	0.1658 ab	0.0029 a	0.1043 ab	0.1368 a	0.0007 ab	0.3359 ab	0.0019 abc	0.0063 a	4.2249 abcd
<i>Trachurus trachurus</i>	0.0001 a	1.2217 a	0.0018 ab	0.0032 a	0.0247 a	0.6398 bc	7.2918 a	0.1551 ab	0.0014 a	0.0030 ab	0.1152 a	0.0007 ab	0.4694 ab	0.0016 abc	0.0097 ab	10.6841 cdefg
<i>Sciaenops ocellatus</i>	0.0002 a	4.7224 abc	0.0008 ab	0.0006 a	0.0290 a	0.3184 ab	3.5802 a	0.3934 ab	0.0040 a	0.0032 ab	0.0498 a	0.0093 b	0.4602 ab	0.0039 bc	0.0192 ab	3.2760 a
<i>Symphodus tinca</i>	0.0005 ab	12.6148 c	0.0021 ab	0.0056 a	0.0258 a	0.2774 ab	4.6088 a	0.6911 ab	0.0023 a	0.0568 ab	0.0340 a	0.0011 ab	0.4107 ab	0.0011 abc	0.0430 ab	4.8118 abcde
<i>Spondyliosoma cantharus</i>	0.0004 ab	3.5180 abc	0.0012 ab	0.0012 a	0.0191 a	0.1823 ab	2.1064 a	0.0922 a	0.0012 a	0.0201 ab	0.0181 a	0.0006 ab	0.5092 b	0.0010 ab	0.0263 ab	6.1340 abcdef
<i>Trachinus draco</i>	0.0004 ab	1.6863 ab	0.0005 ab	0.0033 a	0.0185 a	0.1479 a	1.8022 a	0.0780 a	0.0012 a	0.0472 ab	0.0126 a	0.0004 a	0.3747 ab	0.0009 ab	0.0067 a	5.0897 abcdef
<i>Chelidonichthys lastoviza</i>	0.0001 a	4.1040 abc	0.0006 ab	0.0075 a	0.0345 a	0.1725 ab	1.5771 a	0.3979 ab	0.0017 a	0.0235 ab	0.0308 a	0.0004 ab	0.3895 ab	0.0011 abc	0.0085 ab	5.8579 abcdef
<i>Labrus merula</i>	0.0003 ab	4.5612 abc	0.0028 ab	0.0012 a	0.0182 a	0.2235 ab	3.1480 a	0.9186 ab	0.0019 a	0.0031 ab	0.0171 a	0.0028 ab	0.5395 b	0.0008 ab	0.0174 ab	5.0604 abcdef
<i>Labrus viridis</i>	0.0003 ab	5.2693 abc	0.0008 ab	0.0024 a	0.0478 a	0.2605 ab	3.7136 a	0.3155 ab	0.0010 a	0.0279 ab	0.0073 a	0.0005 ab	0.4256 ab	0.0003 ab	0.0061 a	3.7753 abc
<i>Spicara maena</i>	0.0004 ab	2.6639 abc	0.0031 ab	0.0026 a	0.0284 a	0.2813 ab	24.5842 a	0.4312 ab	0.0026 a	0.0565 ab	0.1144 a	0.0007 ab	0.5171 b	0.0018 abc	0.0182 ab	18.4602 g
<i>Phycis phycis</i>	0.0001 a	11.8097 bc	0.0003 a	0.0004 a	0.0286 a	0.2139 ab	1.8517 a	0.4425 ab	0.0021 a	0.0044 a	0.0585 a	0.0006 ab	0.3963 ab	0.0014 abc	0.0077 ab	3.8294 ab
<i>Pagellus erythrinus</i>	0.0002 a	2.6495 abc	0.0003 a	0.0053 a	0.0169 a	0.2076 ab	2.1229 a	0.1677 ab	0.0013 a	0.0258 ab	0.0191 a	0.0002 a	0.3680 ab	0.0006 a	0.0232 ab	5.0237 abcdef
<i>Scorpaena notata</i>	0.0014 ab	3.6293 abc	0.0042 ab	0.0049 a	0.0403 a	0.2435 ab	4.6572 a	0.3796 ab	0.0033 a	0.2423 b	1.7500 a	0.0013 ab	0.4474 ab	0.0048 abc	0.0163 ab	6.5813 abcdefg
<i>Scorpaena porcus</i>	0.0024 ab	3.5610 abc	0.0083 b	0.0030 a	0.0966 a	0.2945 ab	7.2815 a	0.4662 ab	0.0071 a	0.0958 ab	0.1920 a	0.0013 ab	0.4145 ab	0.0028 abc	0.0321 ab	6.0660 abcdef
<i>Bohus podas</i>	0.0001 a	4.2507 abc	0.0005 ab	0.0001 a	0.0305 a	0.2105 ab	2.8144 a	0.4415 ab	0.0044 a	0.0030 ab	0.0331 a	0.0008 ab	0.3908 ab	0.0016 abc	0.6410 c	7.0531 acdefg
<i>Mullus surmuletus</i>	0.0002 ab	6.7722 abc	0.0010 ab	0.0137 a	0.0358 a	0.3213 ab	33.7140 a	0.3090 ab	0.0140 a	0.0539 ab	0.0371 a	0.0004 a	0.3982 ab	0.0016 abc	0.0274 ab	3.9869 ab
<i>Diplodus vulgaris</i>	0.0005 ab	4.4639 abc	0.0021 ab	0.0098 a	0.0232 a	0.2244 ab	5.5161 a	0.5925 ab	0.0055 a	0.0481 ab	0.0729 a	0.0008 ab	0.4380 ab	0.0022 abc	0.0832 abc	7.8730 abcdefg
<i>Septia officinalis</i>	0.0030 b	11.4781 bc	0.0031 ab	0.0064 a	0.0276 a	1.3903 c	1.5510 a	0.2596 ab	0.0054 a	0.1529 b	0.0504 a	0.0021 ab	0.1978 a	0.0100 c	0.0103 ab	12.3258 efg
<i>Serranus cabrilla</i>	0.0008 ab	1.1554 a	0.0041 ab	0.0006 a	0.0498 a	0.2809 ab	5.7580 a	0.7980 ab	0.0071 a	0.0445 ab	0.0915 a	0.0010 ab	0.4763 ab	0.0022 abc	0.0138 ab	9.0957 abcdefg
<i>Serranus scriba</i>	0.0006 ab	3.3633 abc	0.0016 ab	0.0057 a	0.0324 a	0.3700 ab	4.1159 a	0.2995 ab	0.0023 a	0.1949 ab	0.0419 a	0.0004 a	0.5388 a	0.0012 abc	0.0162 ab	9.5840 bcdefg
<i>Diplodus annularis</i>	0.0010 ab	0.7802 a	0.0024 ab	0.0001 a	0.0321 a	0.2703 ab	4.1644 a	1.4285 b	0.0045 a	0.0807 ab	0.1158 a	0.0007 ab	0.3967 ab	0.0014 abc	0.1282 bc	13.6038 fg

**Table 5** (continued)

Species	Ag	As	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	Sb	Se	Sn	V	Zn
<i>Uranoscopus scaber</i>	0.0009 ab	4.0208 abc	0.0019 ab	0.0038 a	0.0292 a	0.2172 ab	2.0199 a	0.1840 ab	0.0024 a	0.0198 ab	0.0133 a	0.0006 ab	0.3579 ab	0.0008 ab	0.0044 a	4.2240 abcd
<i>Trachinus radiatus</i>	0.0008 ab	1.7261 abc	0.0011 ab	0.0023 a	0.0132 a	0.1752 ab	3.4164 a	0.1537 ab	0.0027 a	0.0310 ab	0.0342 a	0.0008 ab	0.3611 ab	0.0010 abc	0.0097 ab	4.7136 abcde

Different letters (abcdefg) denote significant differences between groups (ANOVA + Tukey HSD test;  $p < 0.05$ )

by European regulations. The THQ and TTHQ values for fish collected near the old Canary asbestos mine were all less than 1 (Table 8). The THQ values for the 9 PTEs associated with fish/mollusk consumption follow the descending order: Se > Pb > Zn > Fe > Cr > Cu > Mn > Cd > Cd. These results suggest that the local population would not be exposed to a potential risk through the consumption of fish. The carcinogenic risk assessment for Pb is 1.2488E-06. This result is within the acceptable range, indicating a carcinogenic risk classified as low (Table 8).

### Discussion

#### Ecological risks

The study of the spatial distribution of PTEs is useful for evaluating possible sources of pollution and identifying contamination hot spots in the study area. The results and multivariate statistical analyses of the potentially toxic elements (PCA, HAC, TEPI, TESVI) made it possible to identify a concentration gradient, a model of bioaccumulation, and the occurrence of different groups, thus reflecting a spatial variation of the contamination. For example, the results of the HAC clearly identified 3 distinct groups, each showing different levels of contamination, with maximum levels of potentially toxic elements at the sites of Albo and Negru (using concentrations in *Posidonia oceanica*) in the immediate vicinity of the Canari mine. In the same way, the maximum values of the concentrations of cobalt, chromium, and nickel, considered tracers of the contamination of the mining activity, were recorded at the sites of Albo and Negru. For example, the average nickel values found on the Albo site were three times higher than those found on the Calvi site. Our study, therefore, indicates that the two most polluted sites (Albo and Negru) are the closest stations to the south of the old Canary asbestos mine. These results are consistent with the literature revealing the presence of comparable high levels of these potentially toxic elements near Canari (Andral et al., 2004; Gosselin et al., 2006; Richir et al., 2015).

Even if the level of contamination remains high, our results show that the PTE concentrations tend to decrease with the distance from the mine, with TEPI values of 1.29 for Albo and 0.99 for Negru. Various studies described the presence of a coastal drift from

**Table 6** Trace Element Pollution Index (TEPI) of fish collected near the former Canari asbestos mine

Species	TEPI	Quality scale	Color scale notation
<i>Scorpaena notata</i>	1.37	High contamination level	
<i>Scorpaena porcus</i>	1.36	High contamination level	
<i>Sepia officinalis</i>	1.27	High contamination level	
<i>Diplodus vulgaris</i>	1.02	High contamination level	
<i>Spicara maena</i>	0.95	High contamination level	
<i>Mullus surmuletus</i>	0.94	High contamination level	
<i>Symphodus tinca</i>	0.88	Medium contamination level	
<i>Serranus cabrilla</i>	0.87	Medium contamination level	
<i>Diplodus annularis</i>	0.85	Medium contamination level	
<i>Serranus scriba</i>	0.82	Medium contamination level	
<i>Sciaena umbra</i>	0.63	Medium contamination level	
<i>Labrus merula</i>	0.57	Medium contamination level	
<i>Scorpaena scrofa</i>	0.56	Medium contamination level	
<i>Trachurus trachurus</i>	0.56	Medium contamination level	
<i>Bothus podas</i>	0.54	Medium contamination level	
<i>Trachinus radiatus</i>	0.52	Medium contamination level	
<i>Uranoscopus scaber</i>	0.51	Medium contamination level	
<i>Chelidonichthys lastoviza</i>	0.50	Low contamination level	
<i>Spondyliosama cantharus</i>	0.46	Low contamination level	
<i>Labrus viridis</i>	0.45	Low contamination level	
<i>Phycis phycis</i>	0.42	Low contamination level	
<i>Pagellus erythrinus</i>	0.40	Low contamination level	
<i>Trachinus draco</i>	0.39	Low contamination level	

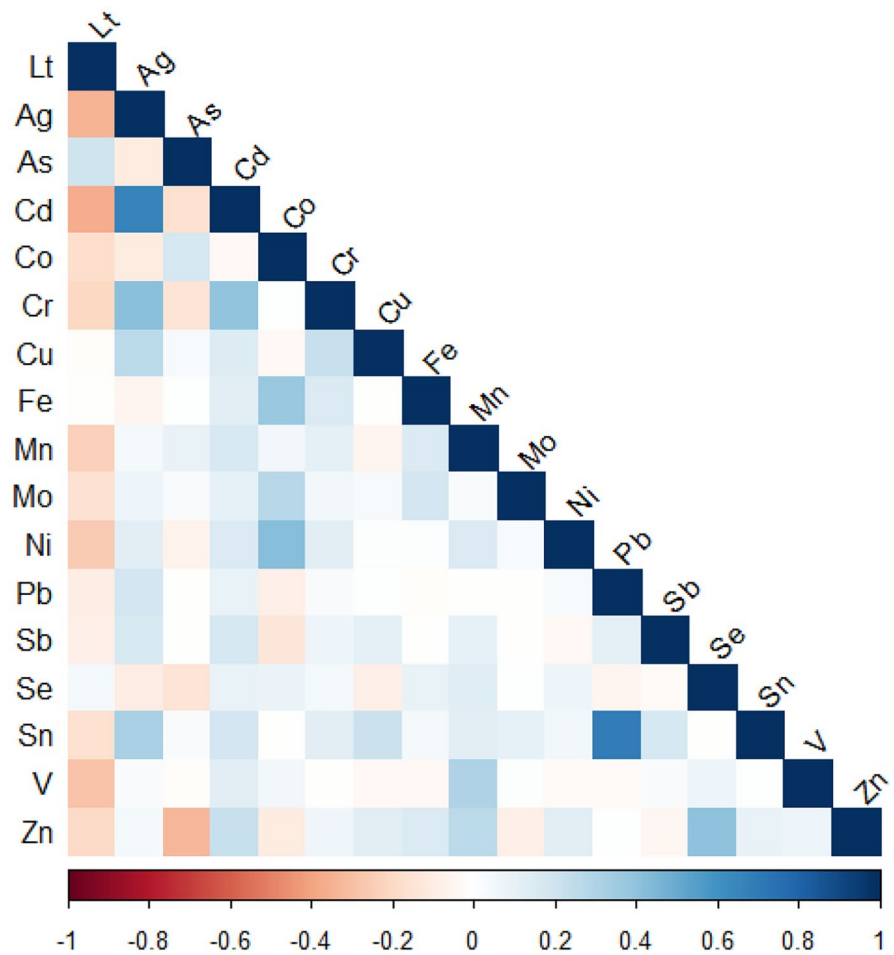
north to south along the west coast of Cap Corse (Bernier et al., 1997; Pluquet, 2006). Therefore, the strong erosion of naturally enriched soils as well as the leaching and drift of waste rocks discharged into the sea during the mining activity (estimated at 11 million tonnes) could explain the higher concentrations found at Albo and Negru than in the other sites situated north of the asbestos mine. Accordingly, the major source of potentially toxic element contaminations in this region seems to be attributed to the natural terrigenous origin and past anthropogenic activities. These results indicate that the former asbestos mine can still be considered, 55 years after its closure, as a major source of Co, Cr, and Ni for

marine ecosystems and still influences the quality of coastal areas today.

On the Canelle, Giottani, and Minerviu sites located north of the old mine, the calculated pollution indices indicate an average/intermediate level of contamination compared to the other sites. The Co, Cr, and Ni concentrations are significantly lower than those at the Albo site but still higher than the levels recorded at the Calvi site. From these elements, we can hypothesize that these three sites can represent the natural background levels of this area of Cap Corse. It has been shown that there is a natural enrichment in this region linked to the local mineralogical and geochemical context (Bouchoucha et al., 2012; Lafabrie et al., 2009). Even



**Fig. 6** Pearson correlation matrix between potentially toxic element concentrations (inter-correlations) and the size (Lt, total length) of fish sampled near the former asbestos mine in Canari



if these sites remain in the zone of influence of the old asbestos mine, we can think that the main way of entering PTEs in these zones would be by natural origin (land inputs, rainwater, rivers, watershed). Finally, the TEPI value obtained for the Calvi site indicates a low

**Table 7** The comparison between recommended values (PTWI) and the estimated weekly intakes (EWI) for fish caught near the former Canari asbestos mine

Element	PTWI (1)	PTWI (70)	EWI
Cu	3500	245000	2.07
Cr	637	44590	0.22
Cd	7	490	3.48
Pb	25	1750	0.01
Zn	7000	490000	47.10

PTWI for 70 kg adult person in micrograms per week per 70 kg body weight. EWI in micrograms per week/70 kg body weight

level of contamination for this site compared to the others. These results are in agreement with the literature, which considers the bay of Calvi and its coastal waters

**Table 8** Target Hazard Quotient (THQ)

Element	Rfd	THQ	CR
Cd	0.001	0.002	
Cr	0.003	0.010	
Cu	0.04	0.006	
Fe	0.7	0.012	
Mn	0.14	0.003	
Pb	0.00357	0.037	1.2488E-06
Se	0.005	0.082	
Zn	0.3	0.024	
<b>TTHQ</b>		0.176	

Total THQ (TTHQ) and carcinogenic risk assessment (CR) linked to the consumption of fish caught near the former Canari asbestos mine (Rfd, oral reference dose)

as having a level of contamination considered to be the lowest in the Mediterranean (Luy et al., 2012; Richir et al., 2013).

In order to better understand the evolution over time of potentially toxic element concentrations in the marine environment, our results were compared with the literature. More particularly with the publication by Lafabrie et al. (2009), which measured the contaminants in the same biological model (adult leaves of *P. oceanica*) on the same site (Albo) during the same season (summer). However, it should be noted that the sampling years are different (2004 and 2019), as are the equipment used and the procedures put in place for the elemental analysis, which was not carried out within the same laboratory. On the Albo site, the concentrations of cobalt and nickel in the leaves decreased, respectively, by  $-33\%$  and  $-41\%$ , between 2004 and 2019. For chromium, the opposite trend is observed with an increase in concentrations of  $+141\%$  in 15 years between 2004 and 2019. Different accumulation models seem to appear depending on the studied elements. This temporal trend could be the result of a marked interannual variation in climatic and environmental factors that influence the inputs of PTEs in coastal systems, as well as their accumulation and/or their bioavailability in *P. oceanica* meadows.

### Human health risks

This study was carried out to assess the risk to human health associated with the consumption of fish in the Canari area. The fish/mollusk collected in this study are commonly consumed by the local population, so monitoring the levels of exposure to these contaminants is of great importance. In this regard, the dataset of the present study provides basic information on the state of potentially toxic element contamination of fish in the Canari area. These data are also a good tool to assess the risk associated with the local consumption of fish in an area enriched in Co, Cr, and Ni.

Chromium is an essential, potentially toxic element that plays a role in the metabolism of glucose, insulin, and lipids (Anderson, 2000). A suboptimal dietary intake of Cr is associated with increased risk factors for diabetes and cardiovascular disease (Mertz, 1993). In our samples, the average Cr concentrations were below regulatory values for human consumption in seafood ( $1 \text{ mg kg}^{-1}$ ). Nickel is not an essential, potentially toxic element, but at low doses, it can be beneficial

in activating certain enzyme systems (Fallah et al., 2011). Chronic absorption of Ni is associated with an increased risk of lung cancer and may lead to neurotoxicity, hepatotoxicity, and nephrotoxicity (Dadar et al., 2016). In our samples, the mean Ni concentrations were below regulatory values for human consumption of seafood ( $0.3 \text{ mg kg}^{-1}$ ). Cobalt is an essential, potentially toxic element in humans as a component of vitamin B12. The element plays an important role in the regulation of blood pressure and is necessary for the proper functioning of the thyroid (Fallah et al., 2011). Its toxicity can affect the thyroid, leading to the overproduction of red blood cells, heart problems, and increased activity in the bone marrow (Medeiros et al., 2012). Cobalt deficiency would also mean vitamin B12 deficiency, leading to muscle weakness, anemia, and nervous problems (Jović & Stanković, 2014). However, it is also recognized as a carcinogen and genotoxic for humans. For Co, there is to date no regulatory threshold established by European legislation concerning the consumption of seafood products. The levels measured in the muscles of fish from Canari are low and similar to those found in other areas of the Mediterranean (Amoussou et al., 2019; Marengo et al., 2018; Minganti et al., 2010).

Regarding the other PTEs measured in the edible tissues of fish, the concentrations were all below the regulatory thresholds and did not reveal any potential risk for human health (Cd, Cu, Fe, Pb, Se, Sn, Zn). In the present study, additional results for the human health risk assessment were obtained from the calculation of several indices and the combination of different approaches (THQ, TTHQ, CR, EWI/PTWI). The daily limit study is considered an excellent tool to assess the balance between the benefits and risks (carcinogenic and non-carcinogenic) associated with eating fish. PTWI is the maximum level of a pollutant to which a person can be exposed throughout their lifetime without appreciable health risk. PTWI values are therefore determined to balance the risk of consumption with the nutritional benefits such as the intake of protein, phosphorus, omega-3 fatty acids, calcium, minerals, and vitamins associated with the consumption of fish. Based on the comparison of the EWI indices with the PTWI values (all lower), the consumption of the fish species in this study did not pose a potential risk to human health. The values of THQ for each element and TTHQ for all elements combined were below the critical value of 1. This result means that the risk of developing

non-carcinogenic diseases from exposure to PTEs is low. Likewise, the results obtained for the calculation of the CR indicate that the probability of developing cancer due to the ingestion of fish from Canari is low (1 chance in 1,000,000,000).

From all these results, which converge towards the same conclusion, it can be said that the consumption of fish in the Canari area does not represent a significant potential risk to human health. As recommended by the American Heart Association or even the Agence nationale de sécurité sanitaire de l'alimentation, de l'environnement et du travail (ANSES), it is recommended as part of a healthy and diversified diet to consume 2 servings of fish per week. This consumption allows optimal coverage of nutrient requirements while limiting the risk of overexposure to chemical contaminants.

However, caution is advised, as the risk assessment for human health is based on a typical population: a 70-kg person, 70 years of a lifespan, and 24 kg of fish consumption per year. It should be noted that the risk for the consumption of these fish has not been assessed on so-called specific populations, e.g., infants, children, pregnant women, and huge consumers of seafood. Thus, we cannot ensure that the same is true and that there is no potential risk for these so-called more vulnerable populations, which would require additional studies. In addition, there are also other sources of exposure to potentially toxic elements, such as the consumption of other foodstuffs (fruits, vegetables, meat, milk, etc.), the inhalation of particles, or even the uptake through the skin, that were not included in this present study and that may have synchronous and cumulative effects.

Several significant positive correlations were found between the different PTEs in the muscles of fish (Co–Ni, Pb–Cd, Ni–Ag, etc.). Inter-elemental relationships could reflect similar assimilation pathways or biochemical regulatory mechanisms between PTEs (Ghribi, 2020). The positive correlations between elements could also reflect a common source of pollution (e.g., Co–Ni and As–Co couple tracers from the former mine) (Yılmaz et al., 2007). The assimilation of PTEs by fish is carried out mainly by the ingestion of suspended particles in the water, ingestion of food, the ion exchange of dissolved PTEs across lipophilic membranes (e.g., gills), and the adsorption on tissue and membrane surfaces. The results showed significant negative correlations between the concentrations of most PTEs (12/16 studied) and the size of the fish. These negative relationships with size could suggest a bioreduction phenomenon. This phenomenon has already

been reported for many fish species in the Mediterranean (Anan et al., 2005). Several mechanisms can explain these negative size–concentration relationships. Younger, smaller individuals often have higher metabolic activity than older, bigger individuals (Storelli et al., 2007). This important metabolic activity in smaller individuals can lead to higher food consumption and also increase the amount of water passing through the gills per unit of time (Canli & Atli, 2003). Since small individuals invest relatively more energy in growth, less energy may be available for detoxification, and this function may be more effective in large individuals (Merciai et al., 2014). This tendency could also be caused by tissue growth being faster than potentially toxic elements intake. If organisms grow faster than they accumulate PTEs, potentially toxic element levels are expected to decrease with increasing body size (Tang et al., 2017). In addition, this hypothesis is consistent with the fact that young/small fish have a lower percentage of adipose tissue than adults/large fish (Braune et al., 1999). Elemental bioreduction is controlled by absorption, elimination, and detoxification, which are highly dependent on metabolism and vary with size/age (Schuster et al., 2019). A negative correlation may also be related to a different absorption rate from the gut and more efficient secretion in the older/taller species. Finally, the size represents an important variable that can reflect ecological, physiological, and morphological changes that could affect the accumulation/reduction of PTEs (e.g., change in energy demand, ontogenetic changes in diet, excretion rate, effect dilution) (Rumbold et al., 2014).

Out of the 23 species, 6 of them (*Scorpaena notata*, *Scorpaena porcus*, *Sepia officinalis*, *Diplodus vulgaris*, *Spicara maena*, *Mullus surmuletus*) showed high contamination levels (TEPI values between 0.94 and 1.37, Table 6). Such differences in the accumulation of potentially toxic elements among species could indicate a phenomenon of bioaccumulation. Several hypotheses can explain the high levels found in these 6 species.

This bioaccumulation is probably due to differences in habitats between the considered species. These 6 species are sedentary, live in close contact with the sediments (e.g., *Scorpaena notata*, *Scorpaena porcus*, *Sepia officinalis*), or are preferably found in the immediate vicinity of *Posidonia oceanica* meadows (*Diplodus vulgaris*, *Spicara maena*, *Mullus surmuletus*). The hypothesis put forward is that their prolonged contact with polluted sediments/meadows during their life would lead to a higher concentration of PTEs in their tissues (Yi et al., 2008). These results are in agreement with the literature, which

indicates that fish living in the upper water column generally have higher concentrations than live fish in the upper water column (Wei et al., 2014). The variations observed between different species can also depend on eating habits. Diet is considered to be the main pathway of bioaccumulation of TE in fish (Zhang & Wang, 2006). The bioavailability of elements from ingested foods is strongly influenced by the nature of the food and varies considerably from one species to another (Metian, 2013). Another hypothesis that could explain these differences in bioaccumulation between fish species could be linked to species-specific physiological characteristics (e.g., genetic markers), including the ability to induce binding proteins such as metallothionein (Damodhar & Reddy, 2012). Organisms have developed a series of mechanisms that control and respond to the uptake and accumulation of PTEs (Bortey-Sam, 2015). These mechanisms include chelation and sequestration through specific proteins (e.g., metallothionein) (Cobbett & Goldsbrough, 2002). Detoxification processes in fish depend mainly on metallothionein, which binds several PTEs together, e.g., Hg, Pb, and Co, facilitating their excretion (Amiard et al., 2006). It would appear that several factors can influence bioaccumulation, depending on the species.

Finally, regarding the 6 most contaminated species (*Scorpaena notata*, *Scorpaena porcus*, *Sepia officinalis*, *Diplodus vulgaris*, *Spicara maena*, *Mullus surmuletus*), given their biological specificities and their life cycles, they can be considered satisfying candidates (bio-indicators) for monitoring the environmental contamination of the coastal waters of Cap Corse.

### Implications for management

The Park, as part of its management plan, wishes to better understand the evolution of contamination in this area in order to initiate, if necessary, management measures adapted to the problem. In perspective, here are some recommendations to try to achieve this goal:

PTE contamination remains a major current concern that can have consequences both for the ecological state of marine ecosystems and on human health. Thus, the pursuit of constant and long-term monitoring of potentially toxic element pollution in the marine ecosystem of the Canari area is strongly recommended.

In order to better understand seasonal variations, it would be appropriate to combine passive biomonitoring (as for this study) with active biomonitoring (e.g.,

caging of mussels). It would also allow for responses at finer temporal and individual scales (e.g., cellular, molecular) by studying stress and exposure biomarkers (damage and defense) such as acetylcholinesterase (AChE), glutathione S-transferase (GST), superoxide dismutase (SOD), or even malondialdehyde (MDA) activity. In addition, in order to correlate these biological responses with environmental factors, it would be desirable to install automated measurement devices at the Canari site (e.g., buoy/fixed multiparameter probe) to acquire data at high frequency and in real time. It would also be interesting to study the quality of the sediments, which are indicators of contamination of the environment because of their capacity to fix pollutants, especially PTEs.

Climatic pressures will have important implications for the marine environment, including the cycle of potentially toxic elements. The Park defines itself as a laboratory and an observatory of climate change, which would allow deploying integrated research that assesses the synergistic and cumulative effects of climate change together with other local anthropogenic pressures.

Greater attention should be paid to trying to reduce the pollution identified in this area at its source, particularly land-based sources. In situ phytoremediation, using plants to restore deteriorated soils, is a promising technology for cleaning up polluted sites (Li et al., 2017). Hyperaccumulative plants are planted to stabilize the soil and to eliminate potentially toxic elements (phytostabilisation, phytoextraction). For example, *Pteris vittata* (L.), the longleaf fern, or *Populus nigra* (L.), the black poplar, are known to take up arsenic in their roots ( $0.2 \text{ mg g}^{-1}$  dry weight) (Briffa et al., 2020). This technique is inexpensive and environmentally friendly. This alternative soil remediation can be coupled with the addition of chelating agents to improve extraction rates. In addition, an effective intervention would be to stabilize the rock waste by covering it with geotextile, which limits erosion and transport to the sea. The substratum can then be revegetated, and with time, the fresh rock surfaces will be oxidized and the release of PTEs will likely be reduced.

Beyond the purely scientific aspects, these results could serve as decision support at the regional level for the definition of long-term public policies. In addition, through awareness-raising, consumers and the local population should be better informed about the potential risks of exposure to contaminants in Cap Corse.



Out of ten asbestos mines in the world (e.g., Canada, China, the USA, Russia, etc.), Canari appears as a particular site for studying the impact of pollution on the sustainability of coastal resources and on the general functioning of marine ecosystems. All the research (current and past) on this site is therefore essential and can serve as a reference state (polluted site) at the Mediterranean level. In view of the unique characteristics of this site and the associated environmental, health, and socio-economic issues, it would be interesting to create a space for exchange to promote the emergence of common synergies between actors/institutions (scientists, managers, state service employees, local elected officials, physicians, associations, etc.). One of the opportunities would perhaps be the creation of a permanent “Canari” commission led by the Park. The objective of this commission would be to put into perspective the scientific data, the means, and the modes of governance existing at the scale of Cap Corse in order to propose suitable tools for the sustainable development of activities in this area. The aim would be, in a more global reflection with a regional coherence, to share the knowledge acquired with all the actors and to help.

## Conclusion

This work provides new and useful information to improve the monitoring of the environmental quality of a region characterized by previous mining activity and to assess the potential risk to human health due to the consumption of fish. This study contributes significantly to the database on chemical contaminants available in different biological compartments in this area. A better understanding of the pressures (of natural and/or anthropogenic origins) and of the processes in action will allow *Parcu Naturale Marinu di u Capi Corsu è di l'Agriate* (PNMCCA, Marine Nature Park) managers to act at the source, reduce deterioration, maintain, or even improve the ecological quality of the coastal waters of Canari. Our results and recommendations could also serve as a model for other asbestos mines still in activity around the world.

**Author contribution** All authors contributed to the study's conception and design. Material preparation, data collection, and analysis were performed by Michel Marengo, Lovina Fullgrabe, and Quentin Fontaine. The first draft of the manuscript was

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**Data availability** The datasets generated during and/or analyzed during the current study are not publicly available but are available from the corresponding author upon reasonable request.

## Declarations

**Ethics approval** Not applicable.

**Consent to participate** Not applicable.

**Consent to publish** Not applicable.

**Conflict of interest** The authors declare no conflict of interest.

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