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ABSTRACT

We report lead (Pb) analyses in juvenile ($n = 37$; mean length = 24.7 ± 2.3 cm) and adult ($n = 16$; mean length = 52.3 ± 9.3 cm) *Centroscymnus coelolepis* Mediterranean deep-sea sharks that are compared to Pb content in bathy-demersal, pelagic and shallow coastal sharks. Median Pb concentrations of *C. coelolepis* muscle (0.009–0.056 wet ppm) and liver (0.023–0.061 wet ppm) are among the lowest encountered in shark records. Stable Pb isotope imprints in adult *C. coelolepis* muscles highlight that most of Pb in *C. coelolepis* is from human origin. Lead isotopes reveal the persistence of gasoline Pb emitted in the 1970s in low-turnover adult shark's muscle while associated liver imprints are in equilibrium with recent pollutant Pb signatures suggesting an efficient pollutant Pb turnover metabolism. The comparison of Pb distribution between adult and juvenile cohorts suggests the role of dietary exposure and possible maternal offloading of Pb during gestation, likely associated to vitellogenesis in this aplacental viviparous deep-sea shark.

1. Introduction

Among toxic metals, lead (Pb) is certainly one of the most pervasive marine pollutants (Boening, 1999; Dell'Anno et al., 2003; Durrieu de Madron et al., 2011; Halpern et al., 2008) due to its long-range transport from combustion processes (metallurgical activities, fossil fuel, and leaded gasoline exhausts), and its short, less than 10 days, atmospheric residence time (Hussain et al., 1998; Nriagu, 1996; Nriagu and Pacyna, 1988; Patterson and Settle, 1987; Poet et al., 1972). Its toxicity is undeniably demonstrated in human (i.e., Canfield et al., 2003; Grandjean, 2010; Lanphear et al., 2005; Needleman, 2004; Tong et al., 1998) and bony fish (Authman et al., 2015; Birge et al., 1981; Burger and Gochfeld, 2005; Danovaro, 2003; Demayo et al., 1982; Heath, 1987; Hodson et al., 1984; Jakimska et al., 2011a; Lee et al., 2019; Mallatt, 1985; Mason, 2013). The former risk has prompted numerous studies in marine ecosystems to define and monitor Pb toxic levels for bony fish consumption (see review in Veron et al., 2021). While fish inhabiting the deep seas, one of the most remote areas of the world, may accumulate high levels of PCBs, organochlorines and metals (Berg et al., 1998; Borghi and Porte, 2002; Escartin and Porte, 1999; Kramer et al., 1984; Sole et al., 2001; Steimle et al., 1990), there is no evidence for atmospherically derived Pb contamination in deep marine fauna although its penetration into the deep ocean is well established (Alleman et al., 1999; Boyle et al., 2014; Veron et al., 1987). Here, we document and investigate Pb content and source in deep-sea selachii (sharks) that are prone to metal absorption and biomagnification owing to their meso to apex trophic position, slow metabolic rates and metal excretion (Adel et al., 2018; Cortes, 1999; Domi et al., 2005; Estrada et al., 2003; Gelsleichter and Walker, 2010; Pethybridge et al., 2010; Shipley et al., 2019).

Sharks are not primary targets for food but rather for cosmetics (squalene) and pharmacology (cartilage) (Bosch et al., 2016; Dent and Clarke, 2015; Dulvy et al., 2014; Mohammed and Mohammed, 2017; Momigliano and Harcourt, 2014; Tiktak et al., 2020) and therefore are

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less investigated than bony fishes for Pb content, most particularly deep-sea species (Bosch, 2015; Corsolini et al., 2014; Gaion et al., 2016; Lozano et al., 2009; Lozano-Bilbao et al., 2018; McMeans et al., 2007; Olmedo et al., 2013; Vas, 1987, 1991; Vas et al., 1993), and mostly in the North Atlantic.

In order to explore pollutant Pb accumulation in deep-sea shark tissues, we choose the bathy-demersal *Centroscymnus coelolepis* (Portuguese dog fish; Bocage and Capello, 1864), one of the deepest living sharks that is distributed worldwide. Its position at the top of food web and estimated long lifespan (Compagno, 1999; Gordon, 1999) that is likely to be as high as the maximum estimated age for a close relative, *Centroselachus crepidater* (54 years old; Irvine et al., 2006) make *C. coelolepis* a suitable candidate as biological sentinel for deep-sea fish contamination. Its year-long aplacental viviparous development constitutes a unique model to investigate pollutant Pb accumulation pathways at different life stages. This shark is mainly a bycatch species in longline and trawl fisheries (Clarke et al., 2002). Meanwhile its slow growth rate, low fecundity and natural mortality make it vulnerable and classified as “Near Threatened” in the IUCN red list (Stevens et al., 2000; Stevens and Correia, 2003). We shall analyze Pb content in both muscle and liver tissues that are of prime interest to assess fish intoxication in relation to metabolic activity (Jakimska et al., 2011b; Kalay et al., 1999; Kojadinovic et al., 2007). Our results will be compared to existing Pb data on Portuguese dog fish and sharks from different habitats, i.e., (i) demersal and bathy-pelagic deep-sea sharks (depth of 1000 m and beyond), (ii) pelagic sharks living in mid-waters and (iii) sharks from shallow coastal waters.

Pollutant Pb origin (from natural or time-transient anthropogenic emissions) shall be assessed with its stable isotopes that we have analyzed for the first time in *C. coelolepis* liver and muscle tissues. Indeed, Pb isotopes (masses: 204, 206, 207 and 208) allow to define the anthropogenic character of Pb owing to differentiated imprints of crustal and ore materials that are inherited from the variations in the initial U—Th content of geogenic reservoirs and the different decay rates of the U—Th parent isotopes (Doe, 1970). This geochemical tracer clearly characterizes the anthropogenic nature and the origin of accumulated Pb in the marine geosphere (Alleman et al., 1999; Angelidis et al., 2011; Boyle et al., 2014; Bridgestock et al., 2018; Kelly et al., 2009; Lee et al., 2014; Noble et al., 2015; Patterson et al., 1976; Stukas and Wong, 1981; Veron et al., 1993, 1998, 1999; Wu et al., 2010). Data on lead isotope ratios are scarce for marine ecosystems (Caurant et al., 2006; Ip et al., 2005; Li et al., 2020; Michaels and Flegal, 1990; Raimundo et al., 2009; Smith et al., 1990, 1992; Spencer et al., 2000; Stewart et al., 2003), and non-existent for sharks. Lead bears insignificant biological or physical isotope fractionation within the marine ecosystem where its isotope imprint is a pertinent marker of anthropogenic sources (Flegal et al., 1987; Michaels and Flegal, 1990; Smith et al., 1990). Therefore, one may expect Pb isotope signatures in *C. coelolepis* to mimic the signature of pollutant Pb sources or a mix thereof. Our field-based approach was conducted in the Mediterranean Sea where osteichthyes (bony fish) display among the highest Pb contents from all investigated oceanic basins (see review in Veron et al., 2021) owing to the proximity to pollutant emission sources from highly industrialized and urbanized areas (Angelidis et al., 2011; Béthoux et al., 1990; Boyle et al., 2014; Copin-Montegut et al., 1986; Durrieu de Madron et al., 2011; Heimbürger et al., 2010, 2011; Migon, 2005; Morley et al., 1997). These Pb emissions are transient in time and isotopic imprint before the 2000s as highlighted in aerosols and corals collected from the Western Mediterranean basin (Migon et al., 2008; Ricolleau et al., 2019). Because pollutant Pb has reached the bottom of the Mediterranean Sea (Angelidis et al., 2011), one may expect deep biota to exhibit Pb contamination that we shall examine in *C. coelolepis* deep-sea shark tissues.

2. Materials and method

2.1. Sampling

Cohorts of juvenile (JC) ($n = 37$; mean length = 24.7 ± 2.3 cm) and adult (AC) ($n = 16$; mean length = 52.3 ± 9.3 cm) Portuguese dogfishes were collected using baited traps at JC (ca. 1900 m depth in 2009) and AC (ca. 2850 m depth in 2001) stations respectively (Fig. 1), in the Western Mediterranean Sea. The JC sharks may be defined as juvenile as their mean length corresponds to the length-at-birth of *C. coelolepis* (i.e., 19–31 cm; Breder and Rosen, 1966; Carrasson et al., 1992; Cox and Francis, 1997; Figueiredo et al., 2008; Girard and De

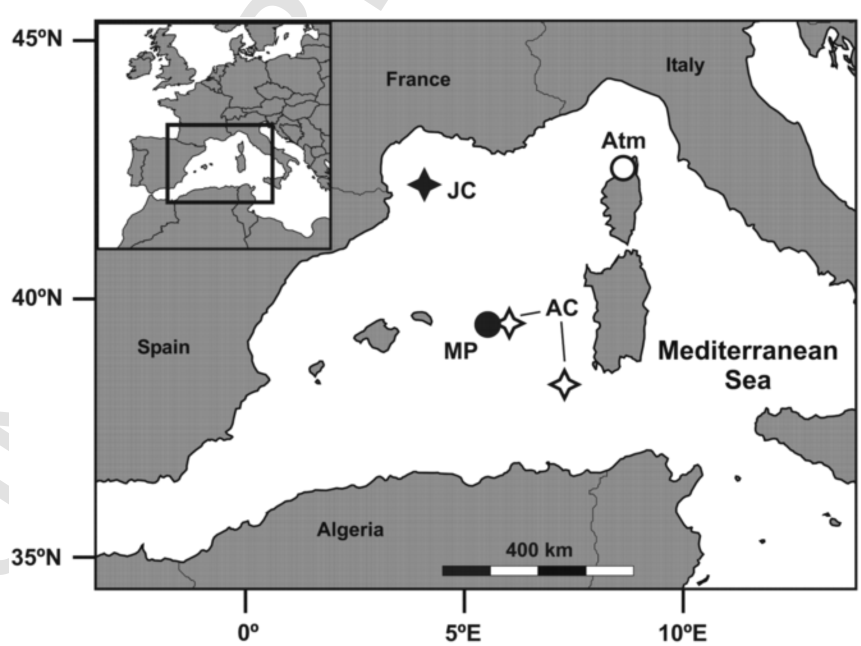

Fig. 1. Sampling sites in the Western Mediterranean Sea. The black and white stars represent JC and AC sampling sites respectively. MP locates marine particle sampling area while Atm (open circle) stands for the location of total atmospheric collection in Corsica.

Buit, 1999; Moura et al., 2011; Torchio and Michelangeli, 1971; Verissimo et al., 2003) while the AC shark median size is within the expected size of adult Mediterranean *C. coelolepis* (Carrasson et al., 1992; Clo et al., 2002; Roule, 1912). Each trap contained squids, horse-mackerel and commercial fish food. Glass spheres (Mod. Vitrovex, 432 mm) and an acoustic release were used to recover the traps. Liver and muscle from each animal were dissected onboard using sterile stainless lancets. All samples were placed into acid-cleaned polypropylene vials and stored at −80 °C until analyses. Samples were freeze-dried before chemical processing for Pb analyses. Geogenic samples were collected to assess the origin of Pb in biologic samples by means of isotopic imprints. Atmospheric (Atm) and Marine Particles (MP, moored particle trap) were collected as part of the ADIOS program in the Algero-Balearic basin (Fig. 1). Monthly bulk atmospheric samples (Atm) were collected in Ostriconi, Corsica from June 2001 to June 2002 while marine particles (MP) were sampled from a 250 m-deep moored trap from April 2001 to May 2002. Sampling procedures for atmospheric and marine particles are described elsewhere (Guieu et al., 2010; Zuniga et al., 2007).

2.2. Lead analyses

C. coelolepis tissues (muscle and liver) of the AC and JC cohorts were digested with a mixture of Merck™ Suprapur© grade HNO_3 and H_2SO_4 in Teflon™ bombs, followed by H_2O_2 addition (Tinggi and Craven, 1996). Lead concentration analyses of the digested AC and JC tissues were performed at the Air and Water Quality Laboratory of the University of the Aegean, Greece (GFAAS Perkin-Elmer 5100ZL Atomic Absorption Spectrometer with Zeeman background) and at the Department of Life and Environmental Sciences of the Polytechnic University of Marche, Italy (by Inductively coupled plasma Mass Spectrometry) respectively. Analytical accuracy was controlled with the use of a Reference Material certified by BCR (CRM 278 mussel's tissue). Stable Pb isotope ratios were determined at GEOTOP (University of Québec in Montréal, Canada) from ca. 50 mg of AC shark tissues that were oxidized in a mixture of distilled Merck™ Suprapur© concentrated HCl, HNO_3 and HF acids before purification on anionic AG1X8 (100–200 mesh) resin (Manhès et al., 1978) before analyses by Multi-Collector Inductively Coupled plasma Mass Spectrometer (MC-ICPMS Micromass Isoprobe). Calibration and mass fractionation were corrected with concurrent thallium analyses and the SRM981 NIST standard. Bulk atmospheric and marine particle samples were processed at CEREGE following the same procedure as at GEOTOP before stable Pb isotope analysis by Thermo Ionization Mass Spectrometry (TIMS Finnigan MAT262). All analytical procedures were conducted in class-100 pressurized clean rooms under laminar flow hoods Total analytical blanks accounted for less than 2% of total analyzed Pb for each processed sample. Lead concentration and isotope ratios for sharks, marine particles (MP) and atmospheric deposition (Atm) are shown in Table S1.

2.3. Literature and statistics

We report Pb levels (ppm wet weight) in muscle and/or livers for sharks living in three different environments and depth ranges, including (DS) demersal-bathy-pelagic deep-sea Sharks (1000 m depth and beyond), (PS) mid-water Pelagic Sharks above 1000 m depth and (SS) Shallow water Sharks in the coastal zones (reefs, sandy beaches, estuaries, mangroves, rocky shores) for comparison (Table S2). Sharks are arranged by genus and species in Table S2 in order to ease inter and intra-species comparison. Results from this study are also presented (in ppm wet weight). Each habitat (DS), (PS) and (SS) comprehends 12 to 14 shark species. Published data were searched using recent peer-reviewed publications from the Web of Science and references therein. In most cases, only papers published in English are considered, and listed in Table S2. Marine records are selected from the world oceans and enclosed seas with the exception of ports to avoid the immediate proximity of polluted ecosystems. Research studies are localized using the Food and Agricultural Organization of the United Nations major fishing areas, subareas, and divisions for the world oceans (FAO-UN, 2021), and the General Fisheries Commission for the Mediterranean (FAO GFCM, 2009) for the Mediterranean Sea. We provide further geographic references such as regional seas, islands and/or countries when available. Studies showing poor accuracy (based on analytical standards), concentration ranges or graphs only, absence of species names or geographic location, and/or whole-body analyses were not considered. Means, number of analyses and dates of sampling are presented for each cohort. The number of analyses for each published mean Pb concentration is taken into account to ascertain a mean/median shark Pb concentration for each ecological habitat (i.e., 2940 and 339 Pb analyses in muscle and liver respectively). In order to match most published Pb content as well as Maximum permissible Limits for fish consumption (MPL), we convert dry mass concentrations into wet ones in Table S2 using: $C_w = C_d.[(100 - H) / 100]$ where C_d and C_w are dry and wet mass concentration (ppm), and H is the percentage of humidity (%) in fish tissues. The conversion factor (CF) is defined as the ratio C_d/C_w. Based on 1477 published CFs, we calculate mean CFs of 4.43 ± 0.24 and 2.64 ± 0.61 for fish muscle and liver respectively (see references in Veron et al., 2021).

Statistical analyses were performed using Past4 statistical package software (Hammer et al., 2001). We present descriptive statistics (arithmetic mean, standard deviation, median, interquartile range) in Table 1. Parametric and non-parametric tests were chosen based on normality test from Skewness index (SKW) and Shapiro-Wilkinson test (SW). Normality and comparison tests use the null hypothesis (H_0) with a given two-tailed probability (p). Failing to reject the null hypothesis is dictated here by $p > 0.05$ (5% chance to detect a false positive). If $p < 0.05$ then we choose the alternative hypothesis, i.e., mean/median differences, non-normal distribution or correlations are statistically significant. Because Pb records from DS, SS and PS cohorts are not normally distributed, medians and Mann-Whitney tests are used rather than means and Student's *t*-test for data comparisons in Tables 1 and 2.

3. Results

3.1. Lead content in adult C. coelolepis (AC)

Lead concentration in *C. coelolepis* determined in this study are presented as ppm dry mass in Table S1 and are converted into wet mass in Tables 1 and S2 to be compared to other published data set (Table S2). We use mean *C. coelolepis* Pb concentrations for comparison to published data that are provided only as means in most articles. Medians are preferred to examine differences between our results and calculated cumulative Pb content in various habitats as explained above. All of the shark Pb medians that are presented in Tables 1 and 2 are within the Maximum Permissible Limits (MPL) that are established to monitor

Table 1
Descriptive statistics ("Mean" and corresponding Standard Deviation "SD", "Median" and Interquartile Range "IQR") of Pb levels (ppm wet weight) in adult (muscle: AC_M and liver: AC_L) and juvenile (muscle: JC_M and liver: JC_L) *C. coelolepis* deep-sea shark from the Western Mediterranean basin (this study). SD and IQR allow to appraise data distribution.

	AC_M	AC_L	JC_M	JC_L
n	15	7	37	37
Mean (sd)	0.077 (0.051)	0.023 (0.004)	0.012 (0.013)	0.109 (0.116)
Med.	0.056	0.023	0.009	0.061
IQR	0.059	0.008	0.009	0.108

Table 2
Descriptive statistics (Mean, Standard Deviation SD, Median, Interquartile Range IQR) of Pb levels (ppm wet weight) in muscle and liver of sharks from various habitats including demersal-bathy-pelagic deep-sea Sharks (DS-1000 m and beyond), mid-water Pelagic Sharks (PS-above 1000 m) and Shallow water Sharks (SS-coastal zones). Medians are calculated from "n" developed mean concentration shown for each shark cohort shown in Table S1.

	DS_M	DS_L	PS_M	PS_L	SS_M	SS_L
n	546	102	169	107	2186	91
Mean	0.11	0.57	0.09	0.48	0.16	0.26
(sd)	(0.10)	(0.67)	(0.01)	(0.49)	(0.12)	(0.29)
Median	0.06	0.26	0.06	0.29	0.15	0.12
P25	0.04	0.08	0.04	0.12	0.14	0.10
P75	0.18	1.26	0.12	1.10	0.16	0.54

fish's toxic metal content and prevent human intoxication, i.e., from 0.3 ppm for the European Union and the FAO/WHO (CAC, 2011; EU, 2015) to 0.5 ppm for Canada, Australia and New-Zealand (CFIA, 2014; FSANZ, 2013). We have categorized shark according to their habitats in spite of potential inter genus-species contrasts in fish Pb content resulting from numerous environmental and biological factors (Burger et al., 2014; Celik et al., 2004; Erasmus, 2004; Jakimska et al., 2011a,b; Kojadinovic et al., 2007; Mathews and Fisher, 2009; Pourang, 1995). Meanwhile, the imprint of contaminated environments on fish Pb content is evidenced with mixed fish species, most particularly for contrasted regions and hot spots in the North Atlantic and the Mediterranean Sea (Celik et al., 2004; Veron et al., 2021). This question has been rarely raised for sharks owing to limited dataset and/or concentrations below detection limits (see Lozano-Bilbao et al., 2018, Turoczy et al., 2000, and references therein). In the most exhaustive investigation of Pb content in demersal sharks from the Northeast Atlantic, Lozano-Bilbao et al. (2018) show that *C. coelolepis* muscle tissues display significantly lower Pb content than other shark species, i.e., *C. cryptacanthus*, *C. uyato*, *C. granulosus*, *C. squamosus*, *D. histricosa* and *D. profundorum*. This discrepancy is also evidenced from the 4 most represented demersal shark genus in our dataset (DS cohort in Table S2) with Pb muscle medians (ppm wet weight) varying from 0.04 (*Centroscymnus*) to 0.07 (*Galeorhinus*), 0.12 (*Centrophorus*) and 0.18 (*Deania*). On the contrary, the two largest records from our dataset display similar Pb muscle medians (0.16 ppm wet weight) for coastal shallow shark's genus (SS in Table S2) *Carcharhinus* ($n = 1526$ analyses) and *Rhizoprionodon* ($n = 608$ analyses). These results may indicate that the influence of environmental vs. biological factors for Pb content in fish fillets may be somewhat driven by the degree of regional Pb contamination, i.e., much higher in littoral than in pelagic and benthic habitats. In order to facilitate further investigation related to inter genus-species discrepancies in Pb content, our shark dataset is presented according to genus and species within each chosen habitat (DS, PS and SS in Table 2).

Muscle Pb mean in *C. coelolepis* adult cohort ($AC_M = 0.077 \pm 0.051$ ppm wet weight; Table 1) is at the highest end of the only published Pb wet mean concentration range in *C. coelolepis* muscles (0.033 ± 0.012, 0.038 ± 0.020 and 0.051 ± 0.033 ppm wet weight; Table S2), measured in 254 specimens from the Macaronesian Islands in the North Atlantic (Lozano et al., 2009; Lozano-Bilbao et al., 2018). These latter references do not mention the size and/or maturity of the analyzed Portuguese dogfishes. Muscle Pb median of the AC cohort (0.056 ppm; Table 1) is similar to that of the deep-sea shark ($DS_M = 0.06$ ppm wet weight; MW $p = 0.60$) and significantly lower than surface shark ($SS_M = 0.15$ ppm wet weight; MW $p < 0.001$) (Table 2). It should be noticed that pelagic (PS) and deep-sea sharks (DS) muscles display similar Pb concentration (Table 2). On the other hand, liver Pb mean in *C. coelolepis* adult cohort ($AC_L = 0.023 \pm 0.004$ ppm wet weight; Table 1) is at the lowest end of the only published Pb wet mean concentration range in *C. coelolepis* liver (0.039 ± 0.030, 0.096 ± 0.107 ppm wet weight; Table S2), measured in 31 specimens from the Azores and Canary Islands (Lozano et al., 2009). AC_L median is significantly lower (MW $p < 0.001$) than DS_L and PS_L median Pb concentrations (Table 2), but significantly higher than SS_L (MW $p < 0.001$). It is difficult to infer why median liver Pb content in coastal surface shark species is lower than in pelagic and deep ones while muscles show the opposite expected trend (median muscle Pb is higher in SS sharks than in DS and PS ones). Biases due to the low number of analyses and limited geographic sampling are possible and these calculated means and medians Pb content in sharks from various habitats should be considered as first order estimates, mostly for cohorts of only 100 or less specimens (Table 2).

3.2. Lead content in juvenile C. coelolepis (JC)

Muscle Pb mean content in *C. coelolepis* juveniles ($JC_M = 0.012 \pm 0.013$ ppm wet weight; Table 1) is at the lowest end of the only published Pb mean concentrations in adult *C. coelolepis* muscles (0.033 ± 0.012, 0.038 ± 0.020 and 0.051 ± 0.033 ppm wet weight; Table S2) (Lozano et al., 2009; Lozano-Bilbao et al., 2018). Muscle Pb median of the JC cohort (0.009 ppm; Table 1) significantly lower (MW $p < 0.001$) than all of the other shark muscle medians from various habitats (Table 2) and from AC_M (Table 1). No muscle Pb concentrations are reported for *C. coelolepis* juveniles at birth. Some data are available for other juvenile sharks 1 to 3 years old that vary between 0.01 and 0.08 ppm wet weight (Kim et al., 2019; Moore et al., 2015; Ong and Gan, 2016). Because of data scarcity, inter species variability and different environmental habitats, one can only infer that mean JC_M are within the lowest range of published data for juvenile sharks. Liver Pb mean content in *C. coelolepis* juveniles (0.109 ± 0.116 ppm wet weight; Table 1) is in the highest end of the two other existing mean liver Pb content in adult *C. coelolepis* (0.039 ± 0.030 and 0.096 ± 0.107; Table S2) measured from 31 specimens caught near the Azores and Canary Islands (Lozano et al., 2009). As for muscles, there is no published Pb content from juvenile *C. coelolepis* at birth. The two other records for juvenile Pb liver content are from immature sharks from shallow coastal waters in the China Sea (*Carcharhinus sorrah*, 0.152 ppm) and the Persian Gulf (*Carcharhinus leiodon*, 0.02 ppm) (Moore et al., 2015; Ong and Gan, 2016). Mean Pb concentration in *C. coelolepis* juvenile liver is significantly higher than in the adult cohort (MW $p < 0.001$, Table 1).

As presented above, mean and median Pb content in muscles of juvenile sharks ($JC_M = 0.012 \pm 0.013$ and 0.009 ppm wet weight) are significantly lower (MW $P < 0.001$) than in adults ($AC_M = 0.077 \pm 0.051$ and 0.056 ppm wet weight) (Table 1). This suggests a more efficient Pb accumulation with growth in low-turnover muscle tissues within adult sharks. The relationship between body length and Pb burden in muscles is not clearly established in sharks owing to the very few existing studies of elasmobranch Pb content at early life stages and/or including all class ages of the same species (Barrera-Garcia et al., 2013; Bosch et al., 2016; Erasmus, 2004; Lopez et al., 2013; Moore et al., 2015). Non-corrected dry weight concentrations of *C. coelolepis* are used to investigate Pb correlations to fish length (cm) (Table S1) to avoid any bias that may be induced by dry-wet correction factor. Muscle from the combined AC and JC cohorts display a moderate positive correlation ($r_{tot} = 0.44$) that is statically significant ($p < 0.001$) (Fig. 2). Spearman test is preferred for the mixed cohorts that are not normally distributed (SW $p < 0.001$) for both length and Pb concentration.

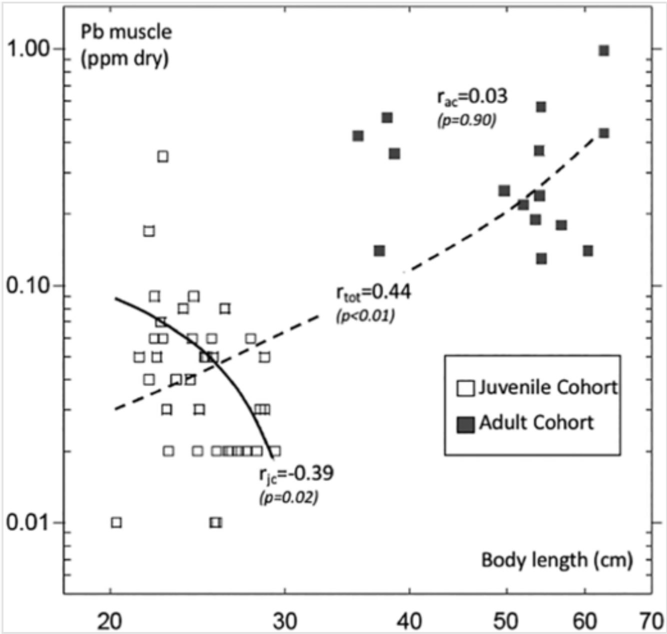

Fig. 2. Relationship between *Centroscymnus coelolepis* body length (cm) and muscle Pb content (ppm dry weight) for the juvenile (open square) and adult (close square) cohorts (see data in Table S1). Spearman correlation coefficients (with corresponding probability *p*) are shown for each cohort (r_{ac} and r_{jc}) and the combined cohorts (r_{tot}).Relationship between *Centroscymnus coelolepis* body length (cm) and muscle Pb content (ppm dry weight) for the juvenile (open square) and adult (close square) cohorts (see data in Table S1). Spearman correlation coefficients (with corresponding probability *p*) are shown for each cohort (r_{ac} and r_{jc}) and the combined cohorts (r_{tot}).

4. Discussion

4.1. Pb origin in adult *C. coelolepis*

The only available Pb isotopic studies in fish tissues were carried out in the Pearl River Estuary in China and in the British Columbia coastline of the North-eastern Pacific Ocean (Li et al., 2020; Ip et al., 2005). Both studies demonstrate the capability of Pb isotopes to source local ambient water Pb imprint either in the heavily contaminated Pearl River Estuary or British Columbia coastline. These field-based investigations were conducted on bony fishes and results could not be directly transposed to sharks that may display different responses to metal exposure owing to their distinct physiology and life cycles. Spencer et al. (2000) used Pb isotopes from fish otoliths (calcium carbonate concretions) to infer local nursery grounds for various fish populations on the basis of local transient pollutant Pb imprints. Here we apply the same isotopic marker to define the extent of pollutant Pb invasion and sources in *C. coelolepis* from the Western deep Mediterranean. We take advantage of the well-monitored transient pollutant Pb imprint in this area both in the atmosphere and seawater that may be compared to shark's isotopic Pb imprints. The relative geographic separation of the Mediterranean Sea regarding the Atlantic Ocean that results from the shallowness of the Gibraltar Strait explains the isolation of the Mediterranean *C. coelolepis* population from its Atlantic counterparts since the Pleistocene (Catarino et al., 2015; Clo et al., 2002). This separation ensures that most pollutant Pb accumulated in these sharks originates from the Mediterranean basin, and therefore could be reasonably compared to monitored regional imprints. Pollutant Pb isotope imprints are transient in time mostly due to the phasing out of leaded gasoline (Bollhofer and Rosman, 2001; Flament et al., 2002; Grousset et al., 1994; Lovei, 1998; Monna et al., 1995; Nriagu, 1990; Petit et al., 2015; Veron et al., 1999). The latter and the decrease of industrial Pb emissions can be accounted for by a decline of Pb input into the Western Mediterranean basin since the 1980s (Migon and Nicolas, 1998; Migon et al., 1993, 2008; Nicolas et al., 1994; Pirrone et al., 1999). Lead isotopes are measured from livers and muscles of the AC specimens and compared to regional imprints both in the atmosphere and at sea in the vicinity of AC sampling sites (Table S2). French urban areas are chosen to best represent the transient pollutant Pb isotopic imprint in the Algero-Balearic basin owing to (1) regional climatic patterns characterized by dominating northerly main wind regimes at sea level in North-western Mediterranean regions (Alhammoud, 2005; Ulbrich et al., 2012), (2) the overwhelming French Pb emissions as compared to other Western Mediterranean countries until the late 1980s (Pacyna and Pacyna, 2000; von Storch et al., 2003) and (3) the apparent restricted deposition of African pollutant Pb emissions in the lower atmosphere within 100 km offshore northern African countries (Guieu et al., 2010). Fig. 3 shows atmospheric Pb imprints ($^{208}Pb/^{206}Pb$ vs. $^{206}Pb/^{207}Pb$) in French urban areas since the early 1980s. These isotopic signatures correspond to mean $^{208}Pb/^{206}Pb$ and $^{206}Pb/^{207}Pb$ ratios from several data sets (see references in Fig. 2 caption) collected during at least a few days (to a few months) in order to display the most representative isotopic means for the 1990s and 2000s (no such long-term isotope imprint is available from the 1980s for which individual data points are shown). These data include imprints of Municipal Solid Combustors (MSWC) from several French cities that do not vary much with time (Fig. 3) and tend to smooth the calculated means for the 1900s and the 2000s. Mean $^{206}Pb/^{207}Pb$ ratios are significantly different from the 1980s (1.110 ± 0.009) to the 1990s (1.140 ± 0.007) and the 2000s (1.153 ± 0.003) (MW $p < 0.001$), underlining as such the well-known transient character of pollutant Pb isotopes in the atmosphere. These imprints fit the INDustrial (IND) and European Standard Lead Pollution (ESLP) lines defined from industrial and car exhausts (Carignan et al., 2005; Cloquet et al., 2006; Haack et al., 2002, 2003; Veron et al., 1999) (Fig. 3). For comparison purpose, we also report in Fig. 3 the 2001–2002 isotopic imprints of Mediterranean marine particles (MP in Fig. 1) (mean $^{206}Pb/^{207}Pb = 1.174 \pm 0.002$) and of the yearly atmospheric deposition at northern Corsica (Atm in Fig. 1) (mass-weighted $^{206}Pb/^{207}Pb = 1.172$) that is calculated from monthly Pb bulk deposition (Guieu et al., 2010) and associated isotopic imprints (Table S2). While both atmospheric and large marine sinking particles display similar isotopic imprints, they differ from the mean urban signature in the 2000s (Fig. 3). This difference is explained by the well-known input of radiogenic natural Pb associated with Saharan dust being transported at high altitude over the African coast to the Mediterranean Sea at the favor of anticyclonic conditions (Bergametti et al., 1989; Dulac et al., 1996; Hamonou et al., 1999; Molinaroli et al., 1993). Despite the phasing out of leaded gasoline and the fact that less than 10% of the aerosols are from anthropogenic origin, pollutant Pb still represents more than 80% of total Pb deposition to the Western Mediterranean in the 1990s and 2000s (Chester et al., 1993; Guerzoni et al., 1999; Guieu et al., 2002, 2010).

Liver (mean $^{206}Pb/^{207}Pb = 1.148 \pm 0.005$) and muscle (mean $^{206}Pb/^{207}Pb = 1.125 \pm 0.006$) isotope imprints of the adult AC specimens are clearly different from the natural crustal Pb imprint (Fig. 3) revealing as such the overwhelming invasion of pollutant Pb in *C. coelolepis* tissues. Lead isotope ratios in AC muscles can clearly be assigned to pollutant Pb from the 1980s, i.e., gasoline Pb, while livers mimic more recent imprints from the late 1990s and early 2000s (Fig. 3) as expected from its detoxifying metabolic activity. The isotopic difference ($^{206}Pb/^{207}Pb$ ratios) between muscle and liver of the same AC sharks (1.2–2.6%) (6 specimens; Table S1) is close to that between French urban isotopic imprints in the 1980s and 1990s (2.6%). The very few stable Pb isotopic atmospheric imprints available from the literature prior to the 1980s do not permit to establish a reliable trend.

However, published $^{206}Pb/^{207}Pb$ ratios from the 1970s (1.11–1.13) are similar to those measured in the 1980s (Chow et al., 1975; Elbaz-Poulichet et al., 1984; Petit et al., 2015) suggesting that a fraction of pollutant Pb in *C. coelolepis* AC muscles may even be as old as the 1970s. Recent direct Pb intake from food is very unlikely to explain the sequestration in muscles of 20 or 30-year-old pollutant Pb. Indeed, in order to ingest this "old" Pb at later life stages, AC sharks should scavenge on preys old enough to have accumulated gasoline Pb previous to its phasing out. However, Mediterranean *C. coelolepis* seldom scavenge and mostly feed on small cephalopods, decapods and bony fishes (Carrasson et al., 1992; Sion et al., 2004) that are not long-lived enough species to have incorporated "pre-phasing out" Pb. We can infer from this isotopic approach that most Pb content in the adult *C. coelolepis* is from anthropogenic origin, with a significant disparity between liver and muscle. The latter reveals that *C. coelolepis* muscles have incorporated a substantial amount of old gasoline Pb emitted in in the 1970s while livers are in isotopic equilibrium with contemporary Pb imprints suggesting a continuous detoxifying metabolism.

4.2. Pollutant Pb accumulation dynamics

Dietary and/or aqueous exposure are the two main pathways for fish uptake of metal which accumulation vary according to species-specific metabolism and habitats, fish life stage and history, metal speciation and bioavailability (see reviews in: Couture and Pyle, 2011; Luoma and Rainbow, 2005; Wang and Rainbow, 2008). Because of peculiar physiological and metabolic aspects (excretion, osmoregulation, organ composition) and life history (longevity, late sexual maturity, low fecundity, apex predatory role), sharks may display specific metal kinetics and bioaccumulation (Bone et al., 1995; Alves et al., 2016; Camhi et al., 1998; Domi et al., 2005; Jeffree et al., 2006, 2015; Mathews and Fisher, 2009; Mathews et al., 2008; Vas et al., 1993) that

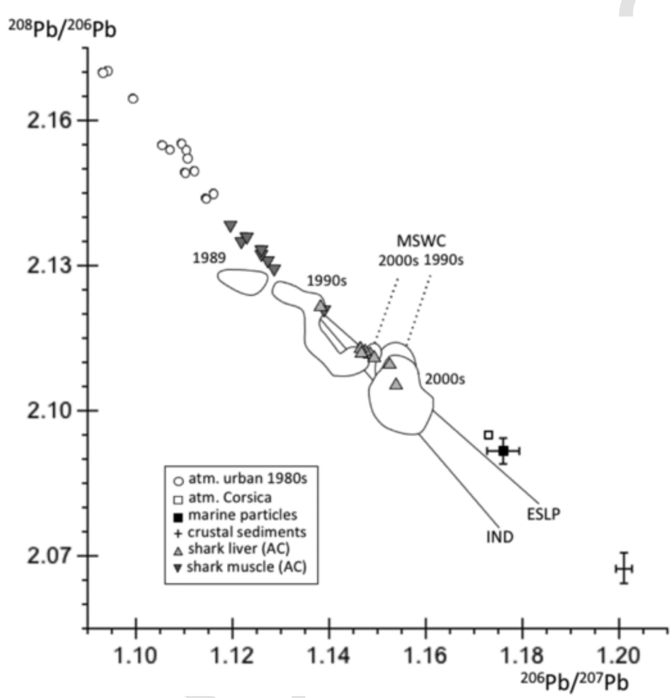

Fig. 3. Comparison of Pb imprints ($^{208}Pb/^{206}Pb$ vs. $^{206}Pb/^{207}Pb$) in *Centroscymnus coelolepis* muscle and liver of AC sharks to those of marine particles and atmospheric deposition (MP and Atm in Fig. 1). Atmospheric Pb isotope ratios of French urban areas (Bollhofer and Rosman, 2001; Carignan et al., 2005; Cloquet et al., 2006; Elbaz-Poulichet et al., 1984, 1986; Monna et al., 1995, 1997; Roy, 1996; Veron et al., 1999; Widory, 2006; Widory et al., 2004) are shown for three decades (1980s, 1990s, 2000s) along with mean non-contaminated sediment imprints of the open Western Mediterranean Sea (Ferrand et al., 1999; Angelidis et al., 2011).

have been examined through modelling, experimental and field approaches (Adams and McMichael, 1999; Branco et al., 2004; Endo et al., 2008, 2015; Hueter et al., 1995; Jeffree et al., 2006; Luoma and Rainbow, 2005; Mathews and Fisher, 2009; Mathews et al., 2008; Pethybridge et al., 2010). Owing to the complexity of factors influencing metal accumulation, we have favored direct Pb analyses from field-caught juveniles and adult sharks. The positive correlation between muscle Pb content and shark length (Fig. 2) suggests a moderate Pb bioaccumulation with time in *C. coelolepis* muscles. Low Pb seawater content and bioavailability in the open Western Mediterranean (Jeffree et al., 2008; Migon et al., 2020) suggest diet exposure as a preferential pathway of Pb accumulation in *C. coelolepis* muscles. This is consistent with the observed increasing levels of metallothioneins with size in *C. coelolepis* (Dell'Anno, unpublished results; Roesdsijadi, 1992). Meanwhile, in highly polluted waters, the placoid scales and high collagen content of the skin shows a high affinity for waterborne Pb in the dogfishes *Squalus acanthias* and *Scyliorhinus canicula* (De Boeck et al., 2010; Eyckmans et al., 2013). This potential *C. coelolepis* dietary Pb uptake should be considered with caution since sampling sites for juveniles and adults were located 300 km away from each other, several years apart. Furthermore, we cannot exclude ontogenetic diet disparities between the two cohorts that live at different depths (Clarke et al., 2001; Gerard and De Buit, 1999; Yano and Tanaka, 1988). Meanwhile, corals collected from the Western Mediterranean basins show (i) no additional decreasing trend in Pb surface seawater content during the 2000s (initially induced by the phasing out of leaded gasoline in the 1980s), and (ii) Pb accumulation disparities of less than 20% in corals from the Gulf of Lions and the Balearic basins during the 2000s (Ricolleau et al., 2019) suggesting no spatial disparities in Pb deposition between the AC and JC sites (Fig. 1). The latter is corroborated by Migon et al. (2008) who found no significant difference in yearly Pb content from Ligurian Sea aerosols during the 2000s. These findings reveal that Pb has reached steady concentrations in the Western Mediterranean atmosphere and seawater during the 2000s. Therefore, it is reasonable to expect Pb content differences between the AC and the JC cohorts to result from biological related issues rather than from recent regional transient disparities in pollutant Pb input within the Western Mediterranean basins.

When considered separately, muscle and length of the AC cohort display no correlation (Spearman $r_{ac} = 0.03$), while the JC cohort shows a weak negative correlation ($r_{jc} = -0.39$, $p < 0.05$) (Fig. 2) that can be explained by a "growth dilution" effect rather than Pb accumulation (Qiu et al., 2011). This result may suggest an ancillary route to Pb uptake, not directly associated with diet or water exposure but possibly with maternal offloading during the two-year aplacental development of the *C. coelolepis* offspring. While matrotrophy (additional nutrition of the embryos) is recognized in viviparous placental sharks (Adams and McMichael, 1999; Frias-Espericueta et al., 2014; Gelsleichter et al., 2007; Hamlett, 1993; Hamlett et al., 2005; Musick and Ellis, 2005; Olin et al., 2014), it remains highly uncertain in *C. coelolepis* that is a yolk-sac ovoviviparous shark, i.e., embryos develop during more than a year on yolk for nutrition (lecithotrophic) before extrusion of fully formed newborns (Figueiredo et al., 2008; Moura et al., 2011; Musick and Ellis, 2005; Yano and Tanaka, 1988). In spite of being defined as aplacental, the inner uterine layer of *C. coelolepis*'s exhibits villosities during gestation suggesting possible maternal offloading (Gerard and De Buit, 1999; Verissimo et al., 2003). For Figueiredo et al. (2008) and Moura et al. (2011), this transfer is limited to water and mineral elements. Matrotrophy is strongly suggested for lamniform aplacental ovoviviparous sharks, *Carcharodon carcharias* and *Alopias vulpinus*, that is associated with lipid transfer from maternal liver during vitellogenesis (yolk formation), histotrophy (feeding from uterine secretions) and oophagy (feeding of the embryos on eggs) rather than continuous maternal supply (Lyons and Lowe, 2013; Mull et al., 2012, 2013; Sato et al., 2016). A radiotracer experiment suggests maternal transfer of metals for the aplacental oviparous shark *Scyliorhinus canicula* during vitellogenesis (Jeffree et al., 2015). While oophagy is not acknowledged for *C. coelolepis* sharks that are considered strictly lecithotrophic i.e., fed on external and internal yolk sacs only, maternal supply associated with vitellogenesis and histotrophy cannot be ruled out. Most particularly, we may hypothesize that the 5–10 times enrichment of Pb content in our juvenile's livers compared to corresponding muscles (Table 1) could result from metal transfer achieved during the constitution of the yolk. Based upon the mean juvenile size (24.7 ± 2.3 cm, see Section 2.1), the JC cohort is considered as "new born" that are not skilled to feed and largely rely on liver for energy source (Endo et al., 2008). Indeed, at birth, elasmobranchs generally possess large livers that provide sufficient energy for early muscle growth, until juveniles are efficient at feeding (Bush and Holland, 2002; Hussey et al., 2009; Lowe, 2002). Therefore, it is reasonable to assume that Pb enrichment in juvenile livers is less likely to be associated with contamination from direct dietary exposure. Other deep-sea juvenile shark cohorts, *Galeus melastomus* (aplacental oviparous) and *Prionace glauca* (placental viviparous), also evidence mean Pb wet concentration 10 to 20 times higher in liver than in muscle tissues (Alves et al., 2016; Gaion et al., 2016).

This study provides insights on the challenging issues related to toxic Pb accumulation source and dynamics in aplacental viviparous deep-sea sharks that remain speculative in our field-based investigation owing to the spatial and temporal differences between juvenile and adult specimens, the lack of physiological data to corroborate our assumptions and the fact that causation cannot be undoubtedly inferred from correlation only. Furthermore, our adult and juvenile *C. coelolepis* cohorts comprised only 34 and 16 specimens, unlikely to represent all maturity stages, rising possible biases for mean calculated Pb concentrations. The latter needs to be further corroborated by interdisciplinary field-based studies that are keys to substantiate and expand experiments and modelling, most particularly for the life cycle and physiological features of deep-sea specimens.

5. Conclusions

The unique collection of juvenile and adult *C. coelolepis* cohorts in the Western Mediterranean Sea allowed us to decipher the Pb burden in this bathy-demersal shark. While its muscle and liver Pb contents are below European Maximum Permissible Limit for fish consumption, and in the lowest encountered Pb concentration range from other shark species from various habitats, pollutant Pb is clearly evidenced in both tissues of the adult cohort where stable Pb isotopes have been measured for the first time. The comparison of the mean Pb isotope imprints in *C. coelolepis* muscles and livers of the latter to recent and transient regional environmental imprints from the Mediterranean atmosphere and marine particles attests the remnant imprint of gasoline Pb from the 1970s–1980s in muscle of the adult cohort. On the contrary, liver metabolic responses appear to efficiently balance pollutant Pb uptake and excretion as shown by its more recent isotope imprint. Lead exposure pathways are explored using the coupling of juvenile and adult cohorts that suggest both dietary intake and potential maternal offloading during early gestational stages for the aplacental ovoviviparous *C. coelolepis* species. This possible exposure of *C. coelolepis* to pollutant metal during its one- to two-year gestation period needs to be further investigated using the combination of several isotope markers and physiological records in juvenile cohorts of viviparous aplacental sharks. The persistence of metals in deep-sea sharks, possibly across generation, should be taken into account when defining strategies and stimulating actions for the conservation and sustainable management of the deep sea.

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Uncited references

Declaration of competing interest

We wish tso confirm that there are no known conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome.

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Descriptive statistics (Mean, Standard Deviation SD, Median, Interquartile Range IQR) of Pb levels (ppm wet weight) in muscle and liver of sharks from various habitats including demersal-bathy-pelagic deep-sea Sharks (DS-1000 m and beyond), mid-water Pelagic Sharks (PS-above 1000 m) and Shallow water Sharks (SS-coastal zones). Medians are calculated from "n" developed mean concentration shown for each shark cohort shown in Table S1.

C. coelolepis tissues (muscle and liver) of the AC and JC cohorts were digested with a mixture of Merck™ Suprapur© grade HNO_3 and H_2SO_4 in Teflon™ bombs, followed by H_2O_2 addition (Tinggi and Craven, 1996). Lead concentration analyses of the digested AC and JC tissues were performed at the Air and Water Quality Laboratory of the University of the Aegean, Greece (GFAAS Perkin-Elmer 5100ZL Atomic Absorption Spectrometer with Zeeman background) and at the Department of Life and Environmental Sciences of the Polytechnic University of Marche, Italy (by Inductively coupled plasma Mass Spectrometry) respectively. Analytical accuracy was controlled with the use of a Reference Material certified by BCR (CRM 278 mussel's tissue). Stable Pb isotope ratios were determined at GEOTOP (University of Québec in Montréal, Canada) from ca. 50 mg of AC shark tissues that were oxidized in a mixture of distilled Merck™ Suprapur© concentrated HCl, HNO_3 and HF acids before purification on anionic AG1X8 (100–200 mesh) resin (Manhès et al., 1978) before analyses by Multi-Collector Inductively Coupled plasma Mass Spectrometer (MC-ICPMS Micromass Isoprobe). Calibration and mass fractionation were corrected with concurrent thallium analyses and the SRM981 NIST standard. Bulk atmospheric and marine particle samples were processed at CEREGE following the same procedure as at GEOTOP before stable Pb isotope analysis by Thermo Ionization Mass Spectrometry (TIMS Finnigan MAT262). All analytical procedures were conducted in class-100 pressurized clean rooms under laminar flow hoods Total analytical blanks accounted for less than 2% of total analyzed Pb for each processed sample. Lead concentration and isotope ratios for sharks, marine particles (MP) and atmospheric deposition (Atm) are shown in Table S1.

We report Pb levels (ppm wet weight) in muscle and/or livers for sharks living in three different environments and depth ranges, including (DS) demersal-bathy-pelagic deep-sea Sharks (1000 m depth and beyond), (PS) mid-water Pelagic Sharks above 1000 m depth and (SS) Shallow water Sharks in the coastal zones (reefs, sandy beaches, estuaries, mangroves, rocky shores) for comparison (Table S2). Sharks are arranged by genus and species in Table S2 in order to ease inter and intra-species comparison. Results from this study are also presented (in ppm wet weight). Each habitat (DS), (PS) and (SS) comprehends 12 to 14 shark species. Published data were searched using recent peer-reviewed publications from the Web of Science and references therein. In most cases, only papers published in English are considered, and listed in Table S2. Marine records are selected from the world oceans and enclosed seas with the exception of ports to avoid the immediate proximity of polluted ecosystems. Research studies are localized using the Food and Agricultural Organization of the United Nations major fishing areas, subareas, and divisions for the world oceans (FAO-UN, 2021), and the General Fisheries Commission for the Mediterranean (FAO GFCM, 2009) for the Mediterranean Sea. We provide further geographic references such as regional seas, islands and/or countries when available. Studies showing poor accuracy (based on analytical standards), concentration ranges or graphs only, absence of species names or geographic location, and/or whole-body analyses were not considered. Means, number of analyses and dates of sampling are presented for each cohort. The number of analyses for each published mean Pb concentration is taken into account to ascertain a mean/median shark Pb concentration for each ecological habitat (i.e., 2940 and 339 Pb analyses in muscle and liver respectively). In order to match most published Pb content as well as Maximum permissible Limits for fish consumption (MPL), we convert dry mass concentrations into wet ones in Table S2 using: $C_w = C_d. [(100 - H) / 100]$ where C_d and C_w are dry and wet mass concentration (ppm), and H is the percentage of humidity (%) in fish tissues. The conversion factor (CF) is defined as the ratio C_d/C_w. Based on 1477 published CFs, we calculate mean CFs of 4.43 ± 0.24 and 2.64 ± 0.61 for fish muscle and liver respectively (see references in Veron et al., 2021).

Lead concentration in *C. coelolepis* determined in this study are presented as ppm dry mass in Table S1 and are converted into wet mass in Tables 1 and S2 to be compared to other published data set (Table S2). We use mean *C. coelolepis* Pb concentrations for comparison to published data that are provided only as means in most articles. Medians are preferred to examine differences between our results and calculated cumulative Pb content in various habitats as explained above. All of the shark Pb medians that are presented in Tables 1 and 2 are within the Maximum Permissible Limits (MPL) that are established to monitor fish's toxic metal content and prevent human intoxication, i.e., from 0.3 ppm for the European Union and the FAO/WHO (CAC, 2011; EU, 2015) to 0.5 ppm for Canada, Australia and New-Zealand (CFIA, 2014; FSANZ, 2013). We have categorized shark according to their habitats in spite of potential inter genus-species contrasts in fish Pb content resulting from numerous environmental and biological factors (Burger et al., 2014; Celik et al., 2004; Erasmus, 2004; Jakimska et al., 2011a,b; Kojadinovic et al., 2007; Mathews and Fisher, 2009; Pourang, 1995). Meanwhile, the imprint of contaminated environments on fish Pb content is evidenced with mixed fish species, most particularly for contrasted regions and hot spots in the North Atlantic and the Mediterranean Sea (Celik et al., 2004; Veron et al., 2021). This question has been rarely raised for sharks owing to limited dataset and/or concentrations below detection limits (see Lozano-Bilbao et al., 2018, Turoczy et al., 2000, and references therein). In the most exhaustive investigation of Pb content in demersal sharks from the Northeast Atlantic, Lozano-Bilbao et al. (2018) show that *C. coelolepis* muscle tissues display significantly lower Pb content than other shark species, i.e., *C. cryptacanthus*, *C. uyato*, *C. granulosus*, *C. squamosus*, *D. histricosa* and *D. profundorum*. This discrepancy is also evidenced from the 4 most represented demersal shark genus in our dataset (DS cohort in Table S2) with Pb muscle medians (ppm wet weight) varying from 0.04 (*Centroscymnus*) to 0.07 (*Galeorhinus*), 0.12 (*Centrophorus*) and 0.18 (*Deania*). On the contrary, the two largest records from our dataset display similar Pb muscle medians (0.16 ppm wet weight) for coastal shallow shark's genus (SS in Table S2) *Carcharhinus* ($n = 1526$ analyses) and *Rhizoprionodon* ($n = 608$ analyses). These results may indicate that the influence of environmental vs. biological factors for Pb content in fish fillets may be somewhat driven by the degree of regional Pb contamination, i.e., much higher in littoral than in pelagic and benthic habitats. In order to facilitate further investigation related to inter genus-species discrepancies in Pb content, our shark dataset is presented according to genus and species within each chosen habitat (DS, PS and SS in Table 2).

Muscle Pb mean in *C. coelolepis* adult cohort ($AC_M = 0.077 \pm 0.051$ ppm wet weight; Table 1) is at the highest end of the only published Pb wet mean concentration range in *C. coelolepis* muscles (0.033 ± 0.012, 0.038 ± 0.020 and 0.051 ± 0.033 ppm wet weight; Table S2), measured in 254 specimens from the Macaronesian Islands in the North Atlantic (Lozano et al., 2009; Lozano-Bilbao et al., 2018). These latter references do not mention the size and/or maturity of the analyzed Portuguese dogfishes. Muscle Pb median of the AC cohort (0.056 ppm; Table 1) is similar to that of the deep-sea shark ($DS_M = 0.06$ ppm wet weight; MW $p = 0.60$) and significantly lower than surface shark ($SS_M = 0.15$ ppm wet weight; MW $p < 0.001$) (Table 2). It should be noticed that pelagic (PS) and deep-sea sharks (DS) muscles display similar Pb concentration (Table 2). On the other hand, liver Pb mean in *C. coelolepis* adult cohort ($AC_L = 0.023 \pm 0.004$ ppm wet weight; Table 1) is at the lowest end of the only published Pb wet mean concentration range in *C. coelolepis* liver (0.039 ± 0.030, 0.096 ± 0.107 ppm wet weight; Table S2), measured in 31 specimens from the Azores and Canary Islands (Lozano et al., 2009). AC_L median is significantly lower (MW $p < 0.001$) than DS_L and PS_L median Pb concentrations (Table 2), but significantly higher than SS_L (MW $p < 0.001$). It is difficult to infer why median liver Pb content in coastal surface shark species is lower than in pelagic and deep ones while muscles show the opposite expected trend (median muscle Pb is higher in SS sharks than in DS and PS ones). Biases due to the low number of analyses and limited geographic sampling are possible and these calculated means and medians Pb content in sharks from various habitats should be considered as first order estimates, mostly for cohorts of only 100 or less specimens (Table 2).

Muscle Pb mean content in *C. coelolepis* juveniles ($JC_M = 0.012 \pm 0.013$ ppm wet weight; Table 1) is at the lowest end of the only published Pb mean concentrations in adult *C. coelolepis* muscles (0.033 ± 0.012, 0.038 ± 0.020 and 0.051 ± 0.033 ppm wet weight; Table S2) (Lozano et al., 2009; Lozano-Bilbao et al., 2018). Muscle Pb median of the JC cohort (0.009 ppm; Table 1) significantly lower (MW $p < 0.001$) than all of the other shark muscle medians from various habitats (Table 2) and from AC_M (Table 1). No muscle Pb concentrations are reported for *C. coelolepis* juveniles at birth. Some data are available for other juvenile sharks 1 to 3 years old that vary between 0.01 and 0.08 ppm wet weight (Kim et al., 2019; Moore et al., 2015; Ong and Gan, 2016). Because of data scarcity, inter species variability and different environmental habitats, one can only infer that mean JC_M are within the lowest range of published data for juvenile sharks. Liver Pb mean content in *C. coelolepis* juveniles (0.109 ± 0.116 ppm wet weight; Table 1) is in the highest end of the two other existing mean liver Pb content in adult *C. coelolepis* (0.039 ± 0.030 and 0.096 ± 0.107; Table S2) measured from 31 specimens caught near the Azores and Canary Islands (Lozano et al., 2009). As for muscles, there is no published Pb content from juvenile *C. coelolepis* at birth. The two other records for juvenile Pb liver content are from immature sharks from shallow coastal waters in the China Sea (*Carcharhinus sorrah*, 0.152 ppm) and the Persian Gulf (*Carcharhinus leiodon*, 0.02 ppm) (Moore et al., 2015; Ong and Gan, 2016). Mean Pb concentration in *C. coelolepis* juvenile liver is significantly higher than in the adult cohort (MW $p < 0.001$, Table 1).

As presented above, mean and median Pb content in muscles of juvenile sharks ($JC_M = 0.012 \pm 0.013$ and 0.009 ppm wet weight) are significantly lower (MW $P < 0.001$) than in adults ($AC_M = 0.077 \pm 0.051$ and 0.056 ppm wet weight) (Table 1). This suggests a more efficient Pb accumulation with growth in low-turnover muscle tissues within adult sharks. The relationship between body length and Pb burden in muscles is not clearly established in sharks owing to the very few existing studies of elasmobranch Pb content at early life stages and/or including all class ages of the same species (Barrera-Garcia et al., 2013; Bosch et al., 2016; Erasmus, 2004; Lopez et al., 2013; Moore et al., 2015). Non-corrected dry weight concentrations of *C. coelolepis* are used to investigate Pb correlations to fish length (cm) (Table S1) to avoid any bias that may be induced by dry-wet correction factor. Muscle from the combined AC and JC cohorts display a moderate positive correlation ($r_{tot} = 0.44$) that is statically significant ($p < 0.001$) (Fig. 2). Spearman test is preferred for the mixed cohorts that are not normally distributed (SW $p < 0.001$) for both length and Pb concentration.

The only available Pb isotopic studies in fish tissues were carried out in the Pearl River Estuary in China and in the British Columbia coastline of the North-eastern Pacific Ocean (Li et al., 2020; Ip et al., 2005). Both studies demonstrate the capability of Pb isotopes to source local ambient water Pb imprint either in the heavily contaminated Pearl River Estuary or British Columbia coastline. These field-based investigations were conducted on bony fishes and results could not be directly transposed to sharks that may display different responses to metal exposure owing to their distinct physiology and life cycles. Spencer et al. (2000) used Pb isotopes from fish otoliths (calcium carbonate concretions) to infer local nursery grounds for various fish populations on the basis of local transient pollutant Pb imprints. Here we apply the same isotopic marker to define the extent of pollutant Pb invasion and sources in *C. coelolepis* from the Western deep Mediterranean. We take advantage of the well-monitored transient pollutant Pb imprint in this area both in the atmosphere and seawater that may be compared to shark's isotopic Pb imprints. The relative geographic separation of the Mediterranean Sea regarding the Atlantic Ocean that results from the shallowness of the Gibraltar Strait explains the isolation of the Mediterranean *C. coelolepis* population from its Atlantic counterparts since the Pleistocene (Catarino et al., 2015; Clo et al., 2002). This separation ensures that most pollutant Pb accumulated in these sharks originates from the Mediterranean basin, and therefore could be reasonably compared to monitored regional imprints. Pollutant Pb isotope imprints are transient in time mostly due to the phasing out of leaded gasoline (Bollhofer and Rosman, 2001; Flament et al., 2002; Grousset et al., 1994; Lovei, 1998; Monna et al., 1995; Nriagu, 1990; Petit et al., 2015; Veron et al., 1999). The latter and the decrease of industrial Pb emissions can be accounted for by a decline of Pb input into the Western Mediterranean basin since the 1980s (Migon and Nicolas, 1998; Migon et al., 1993, 2008; Nicolas et al., 1994; Pirrone et al., 1999). Lead isotopes are measured from livers and muscles of the AC specimens and compared to regional imprints both in the atmosphere and at sea in the vicinity of AC sampling sites (Table S2). French urban areas are chosen to best represent the transient pollutant Pb isotopic imprint in the Algero-Balearic basin owing to (1) regional climatic patterns characterized by dominating northerly main wind regimes at sea level in North-western Mediterranean regions (Alhammoud, 2005; Ulbrich et al., 2012), (2) the overwhelming French Pb emissions as compared to other Western Mediterranean countries until the late 1980s (Pacyna and Pacyna, 2000; von Storch et al., 2003) and (3) the apparent restricted deposition of African pollutant Pb emissions in the lower atmosphere within 100 km offshore northern African countries (Guieu et al., 2010). Fig. 3 shows atmospheric Pb imprints ($^{208}Pb/^{206}Pb$ vs. $^{206}Pb/^{207}Pb$) in French urban areas since the early 1980s. These isotopic signatures correspond to mean $^{208}Pb/^{206}Pb$ and $^{206}Pb/^{207}Pb$ ratios from several data sets (see references in Fig. 2 caption) collected during at least a few days (to a few months) in order to display the most representative isotopic means for the 1990s and 2000s (no such long-term isotope imprint is available from the 1980s for which individual data points are shown). These data include imprints of Municipal Solid Combustors (MSWC) from several French cities that do not vary much with time (Fig. 3) and tend to smooth the calculated means for the 1900s and the 2000s. Mean $^{206}Pb/^{207}Pb$ ratios are significantly different from the 1980s (1.110 ± 0.009) to the 1990s (1.140 ± 0.007) and the 2000s (1.153 ± 0.003) (MW $p < 0.001$), underlining as such the well-known transient character of pollutant Pb isotopes in the atmosphere. These imprints fit the INDustrial (IND) and European Standard Lead Pollution (ESLP) lines defined from industrial and car exhausts (Carignan et al., 2005; Cloquet et al., 2006; Haack et al., 2002, 2003; Veron et al., 1999) (Fig. 3). For comparison purpose, we also report in Fig. 3 the 2001–2002 isotopic imprints of Mediterranean marine particles (MP in Fig. 1) (mean $^{206}Pb/^{207}Pb = 1.174 \pm 0.002$) and of the yearly atmospheric deposition at northern Corsica (Atm in Fig. 1) (mass-weighted $^{206}Pb/^{207}Pb = 1.172$) that is calculated from monthly Pb bulk deposition (Guieu et al., 2010) and associated isotopic imprints (Table S2). While both atmospheric and large marine sinking particles display similar isotopic imprints, they differ from the mean urban signature in the 2000s (Fig. 3). This difference is explained by the well-known input of radiogenic natural Pb associated with Saharan dust being transported at high altitude over the African coast to the Mediterranean Sea at the favor of anticyclonic conditions (Bergametti et al., 1989; Dulac et al., 1996; Hamonou et al., 1999; Molinaroli et al., 1993). Despite the phasing out of leaded gasoline and the fact that less than 10% of the aerosols are from anthropogenic origin, pollutant Pb still represents more than 80% of total Pb deposition to the Western Mediterranean in the 1990s and 2000s (Chester et al., 1993; Guerzoni et al., 1999; Guieu et al., 2002, 2010).

Liver (mean $^{206}Pb/^{207}Pb = 1.148 \pm 0.005$) and muscle (mean $^{206}Pb/^{207}Pb = 1.125 \pm 0.006$) isotope imprints of the adult AC specimens are clearly different from the natural crustal Pb imprint (Fig. 3) revealing as such the overwhelming invasion of pollutant Pb in *C. coelolepis* tissues. Lead isotope ratios in AC muscles can clearly be assigned to pollutant Pb from the 1980s, i.e., gasoline Pb, while livers mimic more recent imprints from the late 1990s and early 2000s (Fig. 3) as expected from its detoxifying metabolic activity. The isotopic difference ($^{206}Pb/^{207}Pb$ ratios) between muscle and liver of the same AC sharks (1.2–2.6%) (6 specimens; Table S1) is close to that between French urban isotopic imprints in the 1980s and 1990s (2.6%). The very few stable Pb isotopic atmospheric imprints available from the literature prior to the 1980s do not permit to establish a reliable trend. However, published $^{206}Pb/^{207}Pb$ ratios from the 1970s (1.11–1.13) are similar to those measured in the 1980s (Chow et al., 1975; Elbaz-Poulichet et al., 1984; Petit et al., 2015) suggesting that a fraction of pollutant Pb in *C. coelolepis* AC muscles may even be as old as the 1970s. Recent direct Pb intake from food is very unlikely to explain the sequestration in muscles of 20 or 30-year-old pollutant Pb. Indeed, in order to ingest this “old” Pb at later life stages, AC sharks should scavenge on preys old enough to have accumulated gasoline Pb previous to its phasing out. However, Mediterranean *C. coelolepis* seldom scavenge and mostly feed on small cephalopods, decapods and bony fishes (Carrasson et al., 1992; Sion et al., 2004) that are not long-lived enough species to have incorporated "pre-phasing out" Pb. We can infer from this isotopic approach that most Pb content in the adult *C. coelolepis* is from anthropogenic origin, with a significant disparity between liver and muscle. The latter reveals that *C. coelolepis* muscles have incorporated a substantial amount of old gasoline Pb emitted in in the 1970s while livers are in isotopic equilibrium with contemporary Pb imprints suggesting a continuous detoxifying metabolism.

Supplementary Figure