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Original Articles

Contaminant signatures and stable isotope values qualify European conger (*Conger conger*) as a pertinent bioindicator to identify marine contaminant sources and pathways

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ABSTRACT

To evaluate the capacity of the European conger (*Conger conger*) as a reliable bioindicator to investigate marine contaminant sources and pathways, the muscles of 24 individuals caught in a semi-enclosed industrialized bay (Gulf of Fos, France) were analyzed for numerous organic compounds, metal elements, and stable isotopes. The contaminant levels were elevated to moderate, which relates to the great anthropic pressure from the surrounding industries and maritime traffic. Stable isotope analyses finely discriminated between three geographical areas and between two age groups. Mercury (Hg) and arsenic (As) concentrations (nd-1.35 $mg.kg^{-1}$ and 5.94–60.1 $mg.kg^{-1}$ wet weight (ww), respectively), as well as chlorination by-products, were elevated in particular areas, identifying specific industrial sources. Levels of Σ_{42}PCB (7.15–28.67 $\mu g.kg^{-1}$ ww), Σ_{16}PAH (3.64–9.48 $\mu g.kg^{-1}$ ww) and Σ_{10}pesticides (1.91–18.42 $\mu g.kg^{-1}$ ww) in fish muscles, as expected, did not differ among sites.

1. Introduction

Contaminants such as trace metals, polycyclic aromatic hydrocarbons (PAHs), polychlorodibenzodioxins and furans (PCDD/Fs), polychlorobiphenyls (PCBs), pesticides, and chlorination by-products are of major concern in marine ecosystems (UNEP/MAP, 2012, 2017; Boudjellaba et al., 2016; OSPAR, 2018). Their harmful effects to marine ecosystems have long been known and the consumption of contaminated marine organisms may lead to human health effects. Even though regulations are applied in the European Union, data from biomonitoring studies are still missing, in particular concerning fish in polluted coastal hot-spots associated to urban or industrial areas (UNEP/MAP, 2012, 2017; OSPAR, 2018).

Bioaccumulation measurements integrate over time the exposure of marine organisms to the bioavailable fractions of the contaminants from the different environmental compartments. Thus, bioaccumulation levels provide useful information to understand and possibly model the exposure pathways (Wang, 2016). It is also obvious that bioaccumulation data is essential to estimate the threat of marine pollution to human health, particularly through the consumption of seafood (OSPAR, 2018).

The European conger has been identified by volunteer citizens living around the industrial zone of the Gulf of Fos (France), as a promising candidate for the biomonitoring of marine pollution. Users of this marine territory, which hosts the main industrial harbor in France and in the Mediterranean Sea, pointed out its sedentary character and its high trophic level guaranteeing an integrated vision of the marine contamination, and potentially a fine spatial resolution to identify the main pollution sources. In the study area, the European conger is consumed in small quantities and has a relatively low commercial value. Its fishery (10–20 kT in the past decade) is essentially located in the Atlantic, and only 8–10% come from the Mediterranean region (FAO, 2019). This “diplomatic” socio-economic aspect qualifies the European conger as bioindicator candidate (Gramaglia and Mélard, 2019). Moreover, the influence from natural variations is minimized in European conger fished in shallow coastal waters, as its sexual

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segregation limits the potential catches to immature female.

Few studies deal with bioaccumulation of contaminants in European conger. Main focus has been on DDT, PCBs and PCDD/Fs (Storelli et al., 2011, 2012), PAH metabolites (Baali et al., 2016), Hg and other metals (Della Torre et al., 2010; Magalhães et al., 2007; Storelli and Barone, 2013). The chlorination by-products accumulation in this species related to seawater concentrations in the Gulf of Fos was also recently investigated in a study by Boudjellaba et al. (2016). Together, the results obtained in these papers further qualifies the European conger as a relevant marine species for biomonitoring. The aim of the current study was to precise the bioaccumulation features of the European conger, including source identification and intra-specific variability to address its reliability as a bioindicator.

2. Materials and methods

2.1. Study area and European conger

2.1.1. The Gulf of Fos

The Gulf of Fos is located between the Rhône river mouth to the West, the Berre Lagoon to the North and the city of Marseilles to the East. It hosts the most important commercial harbor in France and in the Mediterranean Sea (Fig. 1). The steel industry facilities, all located around the Dock #1, are the main source for most metal elements, in particular Fe, Mn, Cr, Ni, Zn (Table S1). PCBs are not expected to be found in the local industrial outlets, while hydrocarbons are released by most activities around the Gulf of Fos. The industrial outlets containing chlorination by-products are mainly located in Dock #1 and #4 and in the East shore, and well described in a previous publication (Boudjellaba et al., 2016). Finally, the wide petrochemical complex located in the East shore is also a major source of Hg, mainly from the use of mercury electrodes. Also, the more diffuse inputs from the Rhône river (1700 $m^3.s^{-1}$ average flow in Arles, 45 km upstream), *via* the part of its plume that curves back into the Gulf when meteorological conditions are favorable, may constitute a major source of contaminants (Ulses et al., 2005).

2.1.2. The European conger

The European conger (*Conger conger*) offers most of the required characteristics regarding the study of contaminant bioaccumulation in its tissues (Haarich, 2011). It is indeed a sedentary fish of high trophic level, which presents sexual segregation patterns (size and depth distributions) and a unique and fatal gestation. It can be assumed that the individuals fished in inshore waters are exclusively immature females (Cau and Manconi, 1984; O'Sullivan et al., 2003; Correia et al., 2009). Considering these biological characteristics, the influence from natural variations is minimized in the European conger. Thus, the contaminant profile of the sampled fish may be representative of the various sources that contribute to the water contamination in the Gulf of Fos.

2.2. Sample collection and preparation

23 specimens of European conger were caught from July to October 2012 in 14 fishing spots (1 to 5 individuals per spot) named "*a*" to "*m*" (Fig. 1). The fishing locations were separated into 3 zones, according to the estimated influence from continental waters and confinement potential (Ulses et al., 2005). They were referred to as "east coast" (sites *a*, *b*, *c*, *d* and *k*), which received the highest marine influence, "west coast" (sites *e*, *j*, *l*, *m* and *n*) representing the inner semi-enclosed part of the Gulf which was more subject to fresh water inputs from the Rhône river and possible stratification, while the "harbor basins" (sites *f*, *g*, *h* and *i*) represented the most confined sites located by the docks and most industrial outlets (Fig. 1). Sampling was carried out using bottom longlines (30–50 hooks and sardine baits), immersed at a depth of 4–12 m depth. Note that no other fish were caught but European congers. The water temperature ranged from 20 to 24 °C during the fishing period.

A total of 23 European congers were caught, which fulfills the number of specimens required for statistics based on individual analyses (Haarich, 2011). Directly after sampling, their total length (l_T, cm) and weight (w_T, g) were recorded. The muscle tissues were removed using sterile single-use scalpels, placed in aluminum foil (organic analyses) or plastic bags (metal elements analyses), and stored at −32 °C. The muscle samples were then freeze-dried at −55 °C and ground in a ball-mill (Retsch MM400, 3 min at 27 Hz) equipped with agate beads

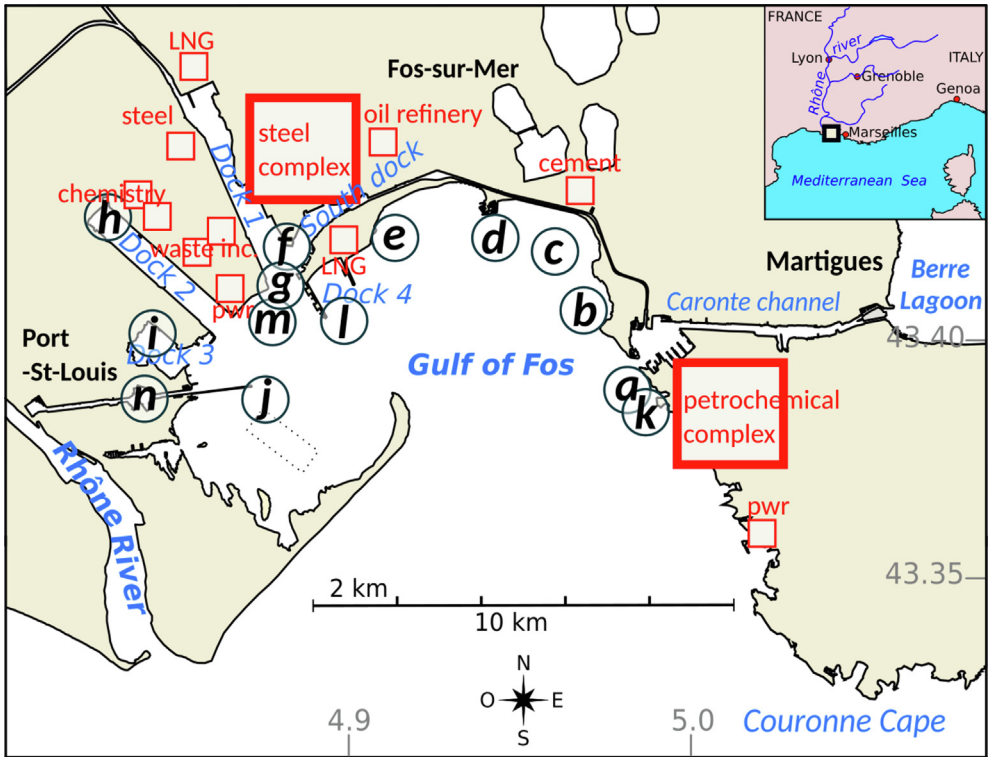

Fig. 1. Localization of sampling sites and situation of the study area in the Gulf of Fos, with main industrial installations (pwr: power plant; waste inc.: waste incineration; LNG: Regazeification plant). In addition, Caronte channel hosts oil and ore terminals, Dock 4 LNG and oil terminals, South dock receives oil tankers, Dock 1 ore and LNG, Dock 2 and 3 host container terminals, and cereal docks stand in the Port-St-Louis channel and Dock 3.

and capsules. The conjunctive tissues were discarded from the homogeneous powder, which was stored in the dark until analysis.

2.3. Chemical analyses

2.3.1. $\delta^{13}C$ and $\delta^{15}N$ values

C and N stable isotope analyses were conducted at the "Littoral Environnement et Sociétés" laboratory (LIENSs – UMR 7266, La Rochelle University/CNRS, France), using an isotope ratio mass spectrometer (Delta V Advantage, Thermo Scientific) coupled to a Thermo Scientific Flash EA1112 elemental analyser. The analytical uncertainty, based on regular acetanilide internal standard measurements, is below 0.1‰ for both $\delta^{13}C$ and $\delta^{15}N$ values (Bustamante et al., 2016). The experimental reproducibility, calculated for the triplicate analyses of 1 mg muscle sample, was below 0.30% for $\delta^{13}C$ and below 0.63% for $\delta^{15}N$.

2.3.2. Metal elements

As detailed in a previous publication (Ratier et al., 2018), about 0.3 g of sample was dissolved in a mixture of nitric acid (7.5 mL, 65% v/v) and hydrochloric acid (2.5 mL, 37% v/v) for the determination of 17 elements (Ag, Al, As, Cd, Co, Cr, Cu, Fe, Hg, Mn, Mo, Ni, Pb, Sb, Sn, Tl, Zn). Microwave mineralization (Ethos advanced μ-wave LabStation) was carried out at 180 °C for 20 min, before ICP-MS analysis (ICAP Q, ThermoElectron).

Mercury (Hg) was prepared separately. Another 0.3 g was dissolved in nitric acid (5 mL, 65% v/v) and 100 μL of a 10 $\mu g\,L^{-1}$ gold solution were added to form a Hg–Au complex to avoid the volatilization of Hg. Samples were kept at room temperature for 72 h before ICP-MS analysis.

Duplicates were prepared and analyzed for all samples. Detection limits ranged from 0.01 to 0.03 $mg\,kg^{-1}$ ww (wet weight) and relative standard deviations (RSD) from 5% to 10%.

2.3.3. PAHs

The 16 PAH congeners defined by the US Environmental Protection Agency (USEPA) priority list, naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (FlA), pyrene (Pyr), benzo(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), benzo(ghi)perylene (Bpe), dibenzo(ah)anthracene (DBA), and indeno(1,2,3-cd)pyrene (IPy), were determined following a long settled and previously described method (Sarrazin et al., 2006; Ratier et al., 2018). Briefly, the samples (~0.5 g) were extracted with 20 mL acetone in an ultrasonic bath (JP Selecta). An aliquot of the supernatant was filtered through a glass microfiber filter (pore size 2.7 μm, Grade GF/D, Whatman). Then, 15 mL of ultrapure water were added, and the solution was passed through a 1-g C-18 cartridge (6 mL volume, Bond Elut, Agilent). The PAH congeners were eluted with 3 × 1 mL acetone followed by 2 × 1 mL methanol. They were finally analyzed by HPLC (Waters 2695 Alliance) equipped with UV and fluorescence detectors (Waters 2487 and 2475) and a C-18 column (Waters 5 μm HAP 4.6 × 250 mm).

Two or three replicates were prepared and analyzed. The PAH congeners detection limits were 0.02 $\mu g\,kg^{-1}$ ww and their average RSD was 24%. The Σ_{16}PAH mean deviations calculated from the replicate analyses of the field samples ranged from 1% to 13%. Additional validation was carried out by analyzing the certified material IAEA 406 (fish homogenate) within the real sample sequences, the results can be found in Table S2.

2.3.4. PCBs and pesticides

The PCB determination focused on 42 congeners including the six PCB indicators (PCBi 28, 52,101,138, 153, and 180) and the 12 dioxin-like congeners (PCB-DL 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189) defined in the European regulations (European Commission, 2011), along with 13 pesticides: lindane, diazinon, heptachlor and epoxydes, aldrin, dieldrin, endrin, endosulfan I and II, and DDT, DDE, and DDD. The analytical method is fully described by Wafo et al. (2006).

Samples (0.5 g) were Soxhlet extracted with hexane and purified with sulfuric acid. The extracts were then submitted to an activated alumina solid phase extraction and eluted with hexane (fraction A). An additional extraction using activated silica and hexane elution (fraction B) was achieved to recover the residuals of eight PCB-DLs (77, 81, 114, 126, 156, 157, 169, and 189) and the pesticides. Fraction A was determined by GC/ECD equipped with a ^{63}Ni detector (HP 6890), and fraction B by GC/MS (Agilent 6890 N). Two or three replicates were prepared and analyzed. The PCB congener detection limits were about 0.01 $\mu g\,kg^{-1}$, and the mean RSD based on the replicate analyses of the field samples ranged from 0.3 to 26% for the Σ_{42}PCB (11.5% in average), and 20.5% in average for individual congeners. Additional validation was carried out by analyzing the certified material IAEA 406 (fish homogenate) within the real sample sequences, the results can be found in Table S2.

2.3.5. PCDD/Fs

The samples of individuals exceeding 100 cm in total length (N = 15) were sent frozen to the Wessling laboratories (Saint-Quentin-Fallavier, France), where 20 g (ww) were ground and homogenized while still frozen. The analysis of the PCDD/Fs was realized as indicated by the EPA Method 1613 (USEPA, 1994), based on HCl digestion in methylene chloride:hexane (1:1, v/v). The organic phase was concentrated and the lipid content was determined. The lipids were removed from the samples with concentrated sulfuric acid, and the organic extract was cleaned-up through activated Florisil column. The samples were finally analyzed by HRGC/HRMS.

2.3.6. Chlorination by-products

As for PCDD/Fs, only the samples of individuals exceeding 100 cm l_T were sent frozen to the Wessling Laboratories for the analysis of 15 chlorination by-products (trihalomethanes, haloacetic acids and halophenols). The analytical procedures and the results are discussed in a paper by Boudjellaba et al. (2016). Only 2,4,6-tribromophenol (TBP) was detected.

2.4. Data analyses

All data analyses, statistical works and modeling were performed using the R software version 3.3 or higher (R Core Team, 2018). Graphical outputs were optimized using the vector graphics editor Inkscape (2018).

Non-linear and (multi)linear regression models were applied to evaluate correlation significance, using the R functions "nls" and "lm", respectively.

The significant differences between mean values were evaluated by the analyse of variance ("aov" R function, outliers omitted) and Tukey post-hoc test ("TukeyHSD" R function), with a confidence level of 0.95.

Hierarchical clustering was performed using the "hclust" R function, using Euclidean distances and ward.d2 criterion applied to standardized data (outliers omitted) (Murtagh and Legendre, 2014).

Outliers removal was strictly limited to the evaluations of significant differences and hierarchical clustering, where it was performed using the "outlier" R package function "rm.outliers" (Komsta, 2011).

3. Results

3.1. Biological parameters

The 23 European conger specimens caught in the Gulf of Fos measured from 70 to 140 cm l_T (average: 110 ± 21 cm l_T) and weighed from 700 to 5 700 g (av.: 2 860 ± 1 640 g), as listed in the Table S3.

Their mean water content was very homogeneous, *i.e.* 74.6 ± 2.4% in average, and the lipid content was 0.43 ± 0.30% of the total weight (Table S3).

The parameters (a = 3.1E-4 and b = 3.39) of the length-weight relationship ($W_T = a \cdot l_T^b$) calculated here fall in the range of those reported in the Western Mediterranean (Giacalone et al., 2010; Crec'hriou et al., 2013) and Atlantic (Fishbase, 2018). This indicates a normal condition index ($K_C = W_T / a \cdot l_T^b$), ranging from 0.69 to 1.28 (average: 0.97 ± 0.09). The age of the sampled fish may be meaningful for bioaccumulation as it may inform on the contaminants integration time. Considering the Von Bertalanffy model ($l_T(t) = l_\infty(1 - e^{-k(t-t_0)})$) and mean literature values for the constants l_∞ = 255 cm, k = 0.058 and t_0 = −1.67 y (Flores-Hernandez, 1990; O'Sullivan et al., 2003; Correia et al., 2009), the European congers sampled here were aged from 4 to 12 y (Table S3).

3.2. Stable isotopes

It has long been demonstrated that information retrieved from N and C stable isotope values recorded in fish tissues supports the description of fish assemblages and food webs (Jarman et al., 1996; Herzka, 2005). The European conger muscles presented elevated $\delta^{13}C$ and $\delta^{15}N$ values of −16.6 ± 0.5‰ and 13.8 ± 0.5‰, respectively (Table S3), underlining their high level in the food chain, compared to recent measurements in other fish samples carried out in the nearby bay of Marseilles (Cresson et al., 2014).

The $\delta^{15}N$ values showed a positive linear trend with total length (R^2 = 0.46, $p < 0.05$) for fish up to 100 cm, while no significant relation between $\delta^{15}N$ and total length was observed for larger specimens (Fig. 2a). This two-step relationship likely reflected a progressive evolution in the young congers diet toward larger preys until they reach approximately 100 cm l_T (*i.e.* 7 years), shifting to settled feeding habits afterwards (Fig. 2a). It was also concomitant with the transition between pre-vitellogenic and early vitellogenic stages estimated to occur at age ≥5 years and l_T > 120 cm (Sbaihi et al., 2001; Correia et al., 2009). Feeding sources and metabolism have a potentially high incidence on contaminants accumulation and elimination, therefore the two size categories will be considered independently.

The contributions of conger total length (as an age proxy) and fishing location, as explanatory variables, were evaluated by multiple linear regression. These two parameters accounted for a total of 58.5% in $\delta^{15}N$ and 35.4% in $\delta^{13}C$ variations. While fish length presented the highest correlations with both isotopic ratios ($p < 0.01$), it was also observed that fishing locations could lead to significant changes in $\delta^{15}N$ values, in particular between harbor basins and east coast ($p < 0.01$) and to a lesser extent between east and west coasts ($p = 0.076$). On the other hand, no difference was observed in $\delta^{13}C$ values according to fishing location.

In order to highlight the shift in isotopic values that could be attributed to spatial distribution, $\delta^{13}C$ and $\delta^{15}N$ were adjusted separately to fish length by covariance analysis. The results, presented in Fig. 2b, confirmed the singularity of the isotopic signatures in the fish samples from the harbor basins compared to those from the west and east shores. The two latter showed slight differences also, in particular in their $\delta^{13}C$ adjusted values. The $\delta^{15}N$ values illustrated greater freshwater inputs in the harbor sites, and to a lesser extent in the west coast. Higher $\delta^{15}N$ isotope values may be associated to more artificially eutrophicated waters (Tu et al., 2014; Matsumoto et al., 2016), which is consistent with the hydrology of the Gulf of Fos. Recent works in the nearby Marseilles Bay also showed that local river inputs induced higher $\delta^{15}N$ and lower $\delta^{13}C$ (Cresson et al., 2014).

3.3. Contaminant levels

All the individual metal elements, PAHs, PCBs and pesticides concentrations are listed in the Supplementary Material Tables S4, S5, S6 and S7, respectively. The chlorination by-products levels are previously reported by Boudjellaba et al. (2016). The contaminants levels are recapped in Table 1 according to fish size, and in Table 2 according to fishing location.

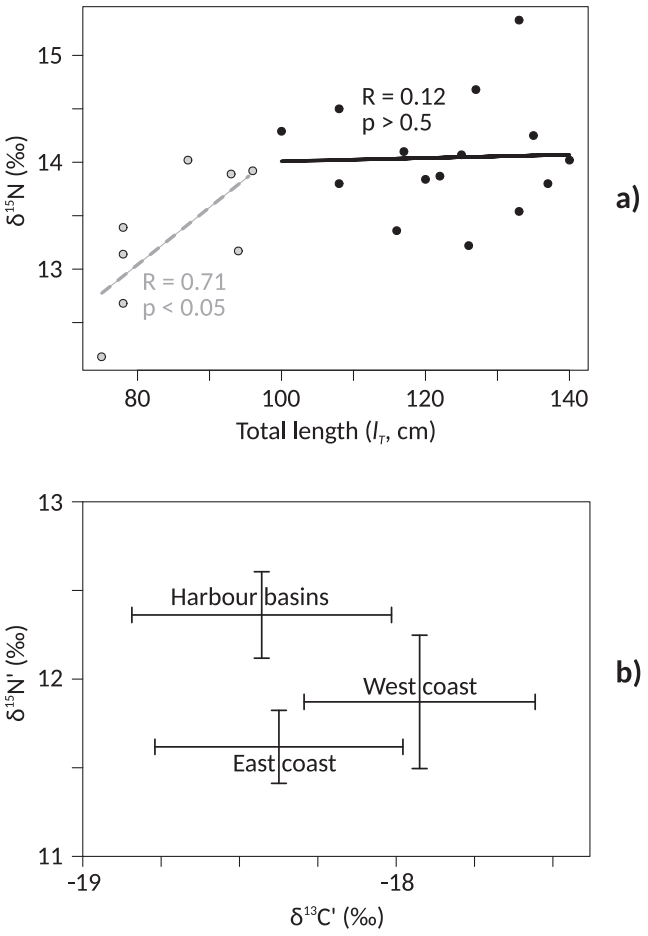

Fig. 2. Relationships between a) $\delta^{15}N$ values and fish total length, and b) mean stable isotope values adjusted to fish length ($\delta^{15}N$' and $\delta^{13}C$') for each zone with 95% confidence intervals.

Globally, metal concentrations (Table S4) were in the range of what measured in *Conger conger* muscles from areas subjected to anthropogenic pressure (Della Torre et al., 2010; Storelli and Barone, 2013). However, reported levels were particularly high for Hg (< 0.1–1.35 mg.kg^{-1} ww), As (5–60 mg.kg^{-1} ww) and Cu (0.4–7.1 mg.kg^{-1} ww). These concentrations, as well as Mo and Mn levels (0.05 and 0.22 mg.kg^{-1} ww in average, respectively), were also higher than in *Anguilla anguilla* muscle samples collected from the nearby Vaccarès Lagoon (Ribeiro et al., 2005). The Hg values were above the European regulations (0.5 mg.kg^{-1} ww (EC, 2006)) in 5 of the 23 samples and all the 21 quantified values were above the background assessment criteria (BAC) established by the UNEP/MAP (2017). Regulated metals Pb and Cd were not detected in any sample, as well as Co, Ag, Sn, Sb and Tl.

The Σ_{16}PAH levels in conger muscles ranged from 3.6 to 9.5 µg.kg^{-1} ww (Table S5, Fig. 3). They are the first report of PAH levels in European conger, and were comparable to other muscle samples from high trophic level fish in the Occidental Mediterranean, 1 to 12 µg.kg^{-1} ww (Perugini et al., 2007; Moraleda-Cibrian et al., 2015). All 16 congeners were detected in all samples. The most abundant congeners were Nap (16–26% of the Σ_{16}PAH) and Ipy (10–23% of the Σ_{16}PAH) followed by Ace and Phe which also exceeded 10% of the Σ_{16}PAH in average. Except for Ipy, this distribution was comparable to what observed in other fish muscle samples collected in the

Table 1
Detected metals (mg.kg^{-1} ww) and organic contaminants concentrations (µg.kg^{-1} ww) according to *Conger conger* size groups (l_T > 100 cm, N = 15, and l_T < 100 cm, N = 8). Evaluation of significant differences between both groups (p, outliers excluded), and correlation coefficients calculated between contaminant levels and total length (R, p < 0.05, N = 23, outliers excluded).

	$l_T \geq 100$ cm	$l_T < 100$ cm	p[a]	R[a]
Metals				
Al	0.09 ± 0.06	0.24 ± 0.05	***	−0.74
Cr	0.48 ± 0.30	1.13 ± 0.70	*	ns
Mn	0.18 ± 0.14	0.28 ± 0.11	*	ns
Fe	2.98 ± 2.66	5.30 ± 2.89	ns	ns
Ni	0.12 ± 0.08	0.40 ± 0.28	**	ns
Cu	3.09 ± 2.16	1.31 ± 1.14	ns	ns
As	22.3 ± 17.1	10.3 ± 4.3	ns	ns
Mo	0.05 ± 0.02	0.08 ± 0.03	ns	ns
Hg	0.32 ± 0.19	0.48 ± 0.44	ns	ns
Zn	3.51 ± 1.84	2.48 ± 0.69	ns	ns
Organic contaminants				
Σ_{16}PAH	7.36 ± 0.90	4.72 ± 0.82	***	0.72
Σ_{42}PCB	27.4 ± 10.7	25.6 ± 8.6	ns	ns
TBP	7.24 ± 2.72	na	na	na
Pesticides				
lindan	0.22 ± 0.07	0.30 ± 0.16	ns	ns
diazinon	0.63 ± 0.26	0.52 ± 0.21	ns	ns
heptachlor	0.31 ± 0.21	0.57 ± 1.49	**	0.46
h. epox. A	0.16 ± 0.07	0.23 ± 0.47	***	0.55
h. epox. B	0.17 ± 0.07	0.70 ± 1.41	ns	ns
endosulfan I	0.73 ± 0.65	1.05 ± 1.67	ns	ns
endosulfan II	2.11 ± 0.70	0.75 ± 0.17	***	0.76
aldrin	0.17 ± 0.14	0.01 ± 0.01	**	ns
endrin	0.13 ± 0.10	0.06 ± 0.13	*	ns
dieldrin	0.31 ± 0.44	0.35 ± 0.44	ns	ns

na: not analyzed.
*** < 0.001; ** < 0.01; * < 0.05; ns: not significant.
[a] p and R values are given excluding outliers.

Mediterranean Sea (Perugini et al., 2007; Moraleda-Cibrian et al., 2015). PCDD/Fs were not detected in any sample, confirming that PCDD/Fs are generally at low levels in marine organisms, often not detectable (Burroni et al., 2009; Storelli et al., 2011).

The 6 indicator PCBs represented 52.5% ± 4.1% of the Σ_{42}PCB determined here. The levels measured in the muscles of congers of the Gulf of Fos, 7.2 to 28.7 µg.kg^{-1} ww (Table S6), are similar to values recorded in European conger from the Ionian and Adriatic Seas, 10 to 25 µg.kg^{-1} ww in average (Storelli et al., 2011, 2012). These concentrations remain elevated, and higher than the ecotoxicological assessment criteria (EAC) established by the UNEP/MAP (2017). As shown in Fig. 4, the PCB profiles were globally dominated by the 6-Cl CB-153, CB-138 and CB-118 (23%, 12% and 5%, respectively), and by the 7-Cl CB-180 and CB-187 (11% and 9%, respectively). They were comparable to what observed in soles from the same area (Dierking et al., 2009) and European congers from the Italian coasts (Storelli et al., 2011, 2012).

Except for DDT and its metabolites, which were never detected, all the insecticides investigated here were measured in all samples (Table S7). The highest levels were recorded for the most contemporary ones, diazinon (0.59 µg.kg^{-1} ww in average) and endosulfan (0.84 and 1.64 µg.kg^{-1} ww in average, for isomers I and II, respectively). The concentrations of the other pesticides, which were all banned in the 90s in France, ranged from 0.11 to 0.54 µg.kg^{-1} ww in average. Except for DDT, the other pesticides are reported for the first time in European conger. However, the concentrations were much lower than those observed in *A. anguilla* in the same region (Ribeiro et al., 2005) and *S. solea* in nearby Gulf of Lions sites (Dierking et al., 2009).

Table 2
Detected metal (mg.kg^{-1} ww) and organic contaminant concentrations (µg.kg^{-1} ww) according to *Conger conger* fishing zone, and evaluation of significant differences between geographic zones (outliers removed and "b" stands for significantly higher).

	West	Harbor	East	diff	p[a]
Metals					
Al	0.14 ± 0.13	0.11 ± 0.08	0.16 ± 0.07	a a a	ns
Cr	0.85 ± 0.73	0.71 ± 0.61	0.52 ± 0.20	a a a	ns
Mn	0.22 ± 0.17	0.24 ± 0.14	0.19 ± 0.11	a a a	ns
Fe	4.7 ± 3.6	3.7 ± 2.9	2.9 ± 2.0	a a a	ns
Ni	0.32 ± 0.29	0.23 ± 0.21	0.10 ± 0.06	a a a	ns
Cu	1.6 ± 1.0	2.4 ± 2.2	3.5 ± 2.4	a a a	ns
As	10.8 ± 3.9	12.8 ± 6.3	32.4 ± 20.5	a a b	* °
Mo	0.06 ± 0.03	0.05 ± 0.02	0.05 ± 0.02	a a a	ns
Hg	0.25 ± 0.09	0.36 ± 0.40	0.46 ± 0.20	a a b	* *
Zn	2.6 ± 1.7	2.9 ± 1.4	4.1 ± 2.0	a a a	ns
Organic contaminants					
Σ_{16}PAH	6.0 ± 1.0	6.3 ± 2.0	7.1 ± 1.5	a a a	ns
Σ_{42}PCB	25.4 ± 10.7	24.5 ± 8.2	31.1 ± 11.7	a a a	ns
TBP	4.5 ± 2.8	8.9 ± 1.0	7.7 ± 2.6	a b ab	°
Pesticides					
lindan	0.22 ± 0.06	0.24 ± 0.16	0.28 ± 0.11	a a a	ns
diazinon	0.66 ± 0.25	0.52 ± 0.18	0.60 ± 0.31	a a a	ns
heptachl.	0.73 ± 1.44	0.14 ± 0.13	0.31 ± 0.21	a a a	ns
h.epox.A	0.26 ± 0.46	0.11 ± 0.07	0.19 ± 0.06	a a b	* °
h.epox.B	0.65 ± 1.42	0.14 ± 0.02	0.26 ± 0.14	a a b	* *
endos. I	1.14 ± 1.65	0.36 ± 0.39	1.05 ± 0.70	ab a b	°
endos. II	1.36 ± 0.78	1.50 ± 0.82	2.10 ± 0.94	a a a	ns
aldrin	0.08 ± 0.11	0.10 ± 0.14	0.18 ± 0.16	a a a	ns
endrin	0.10 ± 0.12	0.07 ± 0.08	0.14 ± 0.13	a a a	ns
dieldrin	0.63 ± 0.54	0.21 ± 0.34	0.10 ± 0.10	b ab a	°

*** < 0.001; ** < 0.01; * < 0.05; ° < 0.1.
ns: not significant.
[a] p and R values are given excluding outliers.

3.4. Bioaccumulation pathways

The bioaccumulation of contaminants in fish tissues depends mainly on waterborne, dietary intake and metabolic elimination (Wang, 2016). Improving the understanding of these pathways is necessary to suggest further interpretations, such as source identification or contaminant environmental fate. Correlations of contaminants with total length may reflect the cumulative effect of exposure with age, while a positive correlation with δ^{15}N would more likely indicate a biomagnification phenomenon (Harmelin-Vivien et al., 2009; Cresson et al., 2014).

The determination of PAHs together with stable isotope values in marine organisms remains scarce in the literature, and their bioaccumulation pathways are still discussed (Takeuchi et al., 2009). Here, Σ_{16}PAH concentrations were strictly referred to on a wet weight basis, as the results on a lipid weight basis did not correlate nor even showed any trend with any biological parameter or stable isotope value. The Σ_{16}PAH was highly correlated with fish total length (R = 0.72, p < 0.001, see Table 2 and Fig. 3), while the relation with δ^{15}N values was not significant (R = 0.40, p = 0.6, see Fig. 3). This indicates that the bioaccumulation of PAHs in the European conger could be related to age rather than trophic level. This finding corroborates the hypothesis that food web biomagnification is not a preeminent pathway for PAH bioaccumulation (Takeuchi et al., 2009; Moraleda-Cibrian et al., 2015; Sun et al., 2016; Ke et al., 2017). The profiles are also dominated by light molecular weight congeners, as found in colocated waters by Guigue et al. (2014). PAH assimilation from water through gills is proposed by other recent studies, and would be consistent here also (Moraleda-Cibrian et al., 2015; Sun et al., 2016; Ke et al., 2017).

Two pesticides, endosulfan II and heptachlor, correlated with total length as well (R = 0.78, p < 0.001, and R = 0.46, p = 0.03, respectively), with strong differences between the ≥100 cm and < 100 cm individuals (Table 1). Significant differences between these two

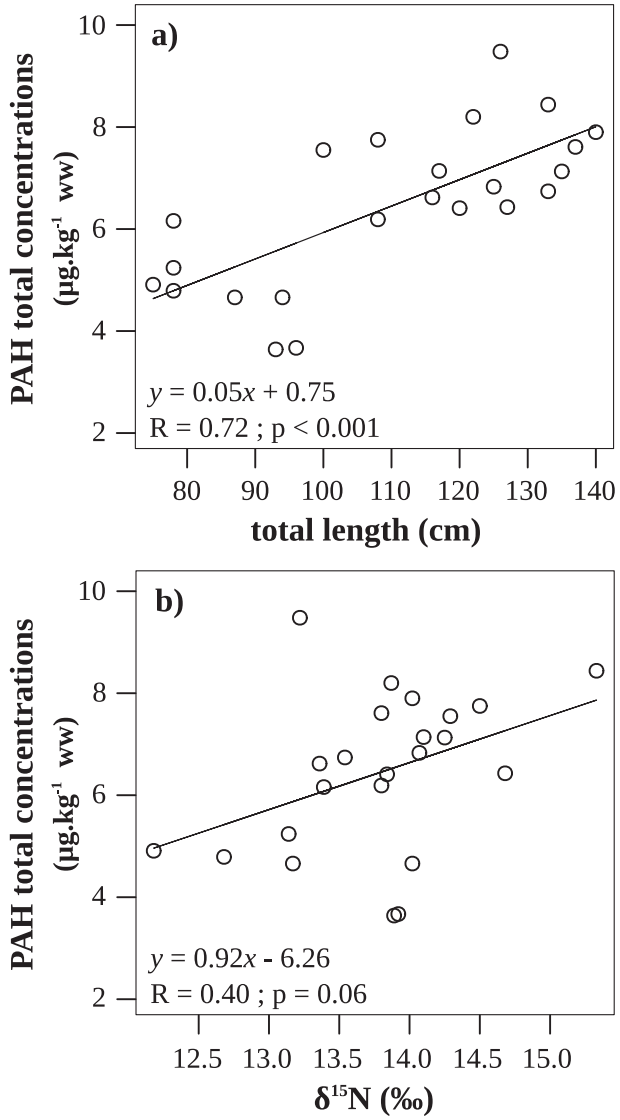

Fig. 3. Relationships between PAH levels in *Conger conger* muscle tissues with a) fish total length, and b) $\delta^{15}N$ values.

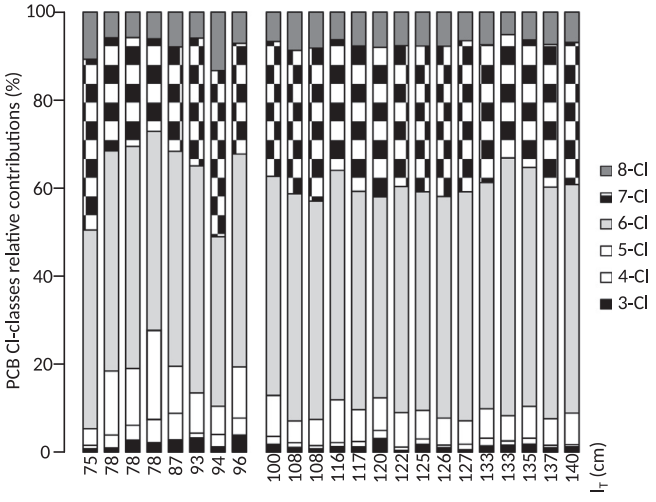

Fig. 4. Relative contributions of PCB according to their chlorine number and *Conger conger* total length (l_T).

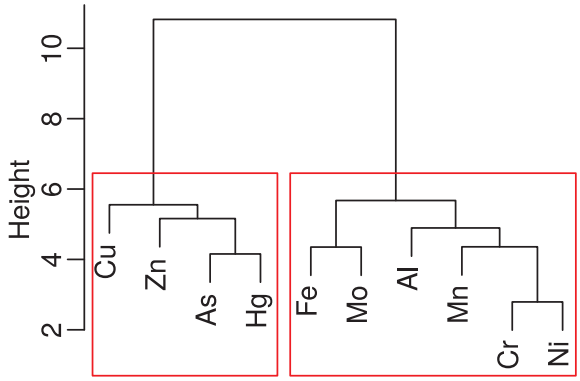

Fig. 5. Hierarchical cluster analysis of metals measured in *Conger conger* muscle tissues from the Gulf of Fos (excluding outliers).

categories were also found for aldrin and endrin concentrations, even though no correlation with total length was observed (Table 1). No significant relation was observed with $\delta^{15}N$ values for any pesticide studied here. In the same manner as PAHs, endosulfan II and heptachlor were likely assimilated by the European conger from water. The differences between the ≥100 cm and < 100 cm specimens may result from a higher metabolism in the younger ones. As aldrin and endrin did not correlate with total length, it was possible that for these pesticides, the effect of metabolism was predominant.

Finally, neither PCBs nor metals presented bioaccumulation or biomagnification patterns. It was quite surprising in the case of PCBs, for which biomagnification is generally significant (Matsuo et al., 2009), but could be explained by the narrow range covered by the $\delta^{15}N$ values in the young and adult *Conger conger* specimens studied here. The geographical location may also become among the prominent factors, even at fine spatial scales as suggested elsewhere (Matsumoto et al., 2016). The concentrations of the elements Al, Cr, Mn and Ni were well correlated (Fig. 5) and significantly higher in < 100 cm specimens (Table 1), possibly reflecting a diet or a metabolic change in relation with their vitellogenic evolution.

4. Discussion

4.1. Insights into the environmental fate of contaminants

In the conger muscle tissues sampled in the Gulf of Fos, the PAH congener profiles were noticeably homogeneous, regardless of total concentrations, $\delta^{15}N$ values, length and spatial categories. This indicated that all fish were probably exposed to similar sources of PAHs. It might also support that the metabolism had a strong homogenization effect toward PAH congeners profiles in *Conger conger* muscle tissues.

In the case of PCBs, the congener profiles were again amazingly homogeneous within conger specimens l_T > 100 cm (Fig. 4), likely reflecting an exposure to the same PCB sources. On the other hand, the distribution of PCB chlorine number classes in young specimens was more erratic, possibly due to the conger diet or metabolic change when they reached l_T = 100 cm (Fig. 4). Also, *Conger conger* travel, in their first 2 years of life, from their hatching area during which they are exposed to different other sources before settling in inshore waters and becoming sedentary. This early life stage may remain critical for some months or years, regarding the PCB accumulation pattern in muscle tissues, as these contaminants are much more persistent in the organisms than PAHs. It is only after 6–7 years, *i.e.* until reaching l_T = 90–100 cm and having spent the majority of their lifetime in their sedentary coastal habitat, that their accumulation will become more representative, thus harmonizing the PCB profiles.

The DDT, which was widely used in the nearby Rhône river delta, and even its metabolites DDD and DDE were not detected, confirming

that the DDT regulation is being effective in France. More contemporary pesticides have shown some interesting trends in the present study, which included the analysis of some metabolites of heptachlor and aldrin. For both, the concentrations of the metabolites or transformation products (epoxydes for heptachlor, and dieldrin for aldrin) were significantly higher in younger fish (Table 2). Heptachlor epoxydes thus represented 81 ± 15% of all heptachlor species in conger when $l_T < 100$ cm, but only 57 ± 14% in older specimens. It was less significant for aldrin, but dieldrin still represented 65 ± 27% in young specimens while only 38 ± 25% when $l_T \geq 100$ cm. This may reflect a higher metabolism of younger specimens or an exposure to heptachlor and aldrin residues that have partly reacted in the environment, and began to decay. Even though the endosulfan-II isomer is expected to have a shorter half-life in sediment due to a preferential microbial degradation than isomer-I and to convert progressively into endosulfan-I in aquatic systems, it generally appears in higher proportions in biological samples (Weber et al, 2010). In the present study, excluding the outlier fished in site "n" which was highly influenced by the cereal marine terminal, the endosulfan I/II isomer ratios were 1.4 and 0.3 in < 100 cm and ≥100 cm individuals, respectively, which is consistent with literature (Weber et al., 2010). This could indicate the major role of metabolism, either in a preferential uptake of isomer-II or a quicker elimination of isomer-I.

Previous studies carried out in the same area report heptachlor epoxides contributions below 5% in the *Solea solea* samples analyzed by Dierking et al. (2009), and from 30% to 95% in *Anguilla anguilla* (Ribeiro et al., 2005). In the present work, dieldrin contributions to (dieldrin + aldrin) in *Conger conger* were in the same order of magnitude than for these *Anguilla anguilla* and sole samples (Ribeiro et al., 2005; Dierking et al., 2009). The endosulfan I/II ratios calculated in *Anguilla anguilla* were also comparable to the present study, and ranged from 0.6 to 1.4 (Ribeiro et al., 2005). This suggested that the degradation of these pesticides was quite limited between 2003 and 2012, and that the lower concentrations in the studied European conger samples are due to inter-specific variability or diffusion of contaminants rather than a decay through chemical or microbial transformations.

4.2. Estimation of contaminant origins

Overall, lower differences were observed between geographical areas than between the two size categories. For instance, PCB total concentrations and congeners profiles measured in the European conger were very similar (Table 2) across the Gulf of Fos. This is in accordance with the Rhône river being the preeminent source of PCBs, dispersing all over the gulf from the North navigation canal and the South back plume (Table S1). The homogeneous PCB distributions were observed on both the wet and the lipid weight basis.

The PAH inputs from the east and west shores of the Gulf of Fos are rather comparable in quantities, and a possibly significant contribution from the Rhône river can also be expected (Table S1). Thus, the uniform concentrations of PAHs in *Conger conger* muscles across the gulf appeared consistent (Table 2). The PAH congeners in the conger samples were also evenly distributed across the gulf, suggesting that the local industrial sources are comparable in the east and west coasts. Petrochemical activities, as well as ore and oil terminals are indeed present in both shores (Fig. 1).

On the contrary, Hg concentrations were significantly higher in the east part of the Gulf of Fos compared to the west coast and to the main harbor basins (Table 2). The preeminence of Hg in fish from the east coast was in accordance with the Lavéra petrochemical complex discharge. It is about the only local source of Hg, and is significant compared to the Rhône river Hg inputs (Table S1), thus leading to a greater exposure of the European conger nearby. The levels of As and to a lesser extend Zn and Cu were also more elevated in the east part of the Gulf of Fos while the other metals were more concentrated in the west shore, although not significantly (Table 2). Also, a hierarchical cluster analysis highlighted that the metal elements could be well separated into two groups, reflecting their distinct origins (Fig. 5). Within these groups, strong correlations between metal concentration values were observed ($R > 0.6$, $p < 0.01$). One group, consisting of Fe, Mn, Cr, Ni and Al, was characteristic of steel industry and ore terminals activities. The Mo was also very specific in the area of a chemical plant located on Dock 2, using Mo in substantial quantities as a catalyzer (Dron and Chamaret, 2015). In the other group, Hg, As and Zn were typical of the petrochemical complex. The distribution of metals in the European conger muscle samples was thus well correlated with the reported industrial discharges (Table S1), except for Zn.

The incidence of specific sources has also been demonstrated for chlorination by-produts (CBPs), by Boudjellaba et al. (2016). The only detected CBP, tribromophenol (TBP), was more concentrated in the European conger from the harbor basins (Table 2), according to the vicinity of LNG terminals and power plants which are the main sources of CBPs into the Gulf of Fos (Fig. 1). TBP was also measured in individuals from sites "a" and "b", possibly reflecting the incidence of the expected chlorinated release from the petrochemical complex.

Finally, the European conger muscle tissue sampled in site "n", by the cereal terminals, presented extreme pesticide concentrations, in particular for heptachlor and endosulfan (Table S7). This revealed that the proximity of the cereal terminals, which use various pesticides to control the cereals integrity in storage silos, led to a pesticides pollution hotspot in the Port-Saint-Louis canal. Apart from this outlier, the heptachlor and its metabolites, as well as endosulfan, were surprisingly more concentrated in the East samples (Table 2).

The study of the contaminant concentrations at a fine spatial resolution in an industrialized bay, exposed to multiple local releases but also to significant remote inputs, revealed some contamination hotspots, specific pollution sources and some more diffuse influences. These observations were well correlated with our knowledge of the pollution sources, and highlight the capabilities of marine biomonitoring with the European conger in environmental assessment issues.

4.3. Health implications

The contaminant levels measured in edible fish tissues are generally compared to toxicological thresholds, that are computed into tolerable daily intakes (TDI) in order to take into account the local fish consumption habits (Storelli et al., 2011, 2012). Here, the daily intakes were calculated based on the TDI values reported in Table S8, for a 70 kg adult and considering a fish consumption of 26.2 $g.day^{-1}$ (France mean value in 2006–07 (ANSES, 2009)). It was also considered the hypothesis that inorganic As represents 0.41% ww in conger muscles, as observed by Leufroy (2012) for most fish species. Also, total Hg was considered rather than methyl-Hg, but it should be noted that the latter is generally known to account for 80–100% ww (Magalhães et al., 2007; Pethybridge et al., 2010). However, the methyl-Hg equivalent can be simply calculated by applying a 2.5-fold conversion, which corresponds to the ratio between total and methyl-Hg TDI established by the JECFA (2018). As PAHs do not benefit from a global TDI, benzo (a)pyrene was considered.

The chemical contaminants of main health concern found in the European conger muscles from the Gulf of Fos were PCBs and Hg (Fig. 6). In average, 26.5% and more than 20% of the tolerable intake for PCBis and Hg, respectively, were brought by the only consumption of fish (considered here as strictly European conger) muscle from the Gulf of Fos. When considered as methyl-Hg, the fish contribution to the Me-Hg tolerable intake rises up to more than 50%. To a lesser extent, other chemical contaminants should be still carefully surveyed, in particular, As, Ni, PAHs, and lindane. The health risk calculated for each size category was comparable (Fig. 6), but there was a strong geographical contrast. The potential intake of Hg and As from European conger consumption was 3-times higher in the east coast than in the west coast and the intake of PCBis was also slightly higher in the east.

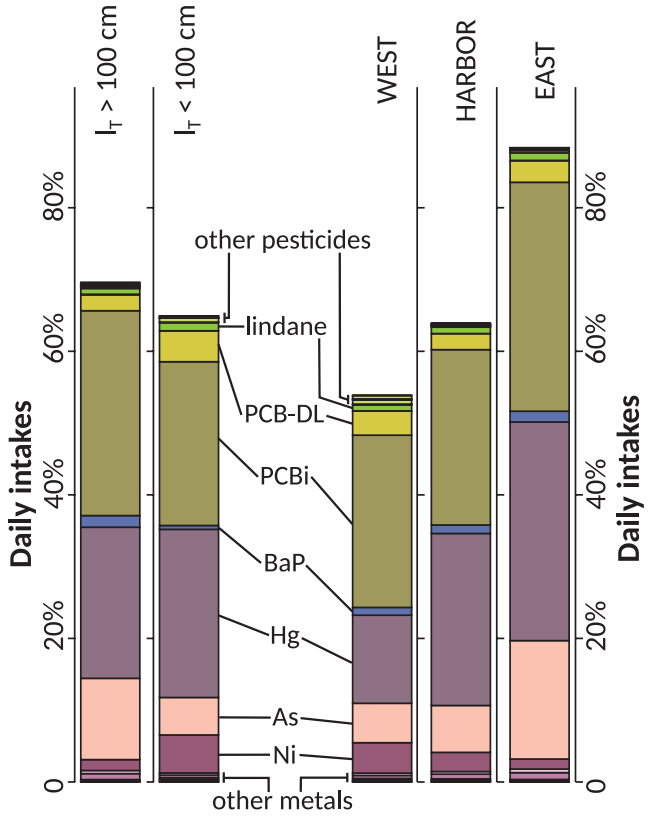

Fig. 6. Stacked daily intakes (%) according to the total length (l_T) and living area of the *Conger conger* sampled (inorganic As = 0.41% ww of total As; Hg = total Hg; BaP = benzo(a)pyrene).

These results show how the European conger, thanks to its biological characteristics and the study of the stable isotope values, represents a powerful tool to assess the health risk at a fine spatial resolution.

5. Conclusions

This study investigated the potentialities of *Conger conger* as a marine bioindicator for chemical contamination at a fine scale. The analysis of the stable isotope composition along with biological parameters did succeed into separating two total length (proxy for age) categories and three distinct areas at a fine spatial resolution.

From this point, and considering the sedentary behavior of the European conger at this life stage, it has been possible to characterize the contamination of the environment by Hg and As elements from petrochemical activities and chlorination by-products sources in the east coast and in the harbor basins of the Gulf of Fos, respectively. To a lesser extent, steel industry markers such as Fe, Mn, Cr and Ni were higher in the conger muscle tissues from the west and the harbor basins, consistently with the location of a major steel complex. On the other hand, it was shown that the bioaccumulation of pesticides and PCBs was mainly due to more remote inputs brought by the Rhône river. Finally, PAHs are probably released by most industrial sources, and were thus quite homogeneous in the samples.

Additionally, insights into the bioaccumulation pathways were pointed out, in particular the hypothesis that PAHs and several pesticides accumulation was not conspicuously due to food web biomagnification, but occurred more probably through the assimilation from water. The decay of several pesticides was also shown through the comparison of the two total length groups, but the narrow range covered by the $\delta^{15}N$ values within the extent of 75–140 cm conger specimens did not reveal the biomagnification expected for PCBs and possibly some metal elements.

These findings offered key elements to assess the detailed exposure of the ecosystem and the threats induced by fish consumption. This study showed more generally the great bioindication abilities offered by the European conger, and indicated that focusing on specimens > 100 cm guarantees an even higher level of independence towards natural fluctuations. These abilities will be a considerable asset when applied elsewhere and at broader scales.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecolind.2019.105562.

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Contaminant signatures and stable isotope values qualify European Conger (Conger conger) as a pertinent bioindicator to identify marine contaminant sources and pathways

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Supplementary material

Table S1. Estimated industrial contaminant releases in the Gulf of Fos (tons.y^{-1} based on 2005 and 2007 regulatory thresholds for the west and east harbor, gathered by BURGEAP, 2008 and 2011, respectively), and fluxes measured in the Rhône river (tons.y^{-1}, 2011-12 averages) at the Arles station (45 km upstream, Launay, 2014; OSR, 2015).

	Rhône riv.[a]	West harb.	East harb.
Al	na	0.17	0.99
Cr	442	0.19	0.01
Mn	na	3.01	ns
Fe	na	20.7	0.06
Co	69	0.04	ns
Ni	133	0.28	0.04
Cu	100	0.28	0.89
Zn	382	7.48	0.55
As	na	0.03	0.01
Cd	1.3	0.01	0.01
Hg	0.45	ns	0.02
Pb	110	0.31	0.11
PCB_i	0.06	ns	ns
Hydrocarbons	10.8[b]	33.2	24.2
CBP	na	10[c]	2.7[d]

na: not available; ns: not significant; CBP: chlorination by-products

[a] Flux reaching the study area (Gulf of Fos) is estimated to be at most, a few percents [Bouloubassi et al., 2012]

[b] PAH-12 in 2002 [Bouloubassi et al., 2012]

[c] bromoform in 2014 [Boudjellaba et al., 2016]

[d] AOX + active Cl [BURGEAP, 2011]

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Table S2. Validation results (µg.kg^{-1} dry weight) for the analysis of the certified material IAEA 406 fish homogenate: certified values and 95% confidence intervals, experimental mean values and standard deviations

PAH (N=2)

	Nap*	Acy*	Ace*	Flu*	Phe*	Ant*	FlA*	**Pyr**	BaA*	Chr*	BbF*	BkF*	BaP*	DBA*	BPe*	Ipy*
Cert.	*24*	*8.0*	*1100*	*na*	*22*	*19*	*4.9*	***4.5***	*0.81*	*2.3*	*2.3*	*2.8*	*2.9*	*na*	*0.78*	*na*
Cert. 95% CI	*16 - 440*	*1.1 - 26*	*0.29 - 2300*	*na*	*8 - 220*	*15 - 43*	*4.0 – 9.6*	***2.1 – 9.1***	*0.24 – 0.91*	*2.2 – 4.4*	*0.78 – 9.5*	*0.38 - 130*	*0.62 – 3.0*	*na*	*0.28 – 1.3*	*na*
Meas.	28.7	6.1	20.2	7.9	13.0	21.0	7.5	**6.4**	0.88	3.1	5.3	58.7	2.6	nd	0.49	1.8
Meas. SD	4.7	1.3	3.0	0.6	3.8	1.6	1.5	**1.4**	0.46	0.8	1.2	4.5	na	nd	0.11	0.3

PCB (N=4)

	CB 28	**CB 52**	**CB 101**	CB 118*	**CB 138**	**CB 153**	**CB 180**
Cert.	**0.57**	**1.3**	**3.1**	2.5	**4.0**	**3.7**	**1.2**
Cert. 95% CI	**0.43 – 1.3**	**1.0 – 2.2**	**2.2 – 3.4**	1.9 – 3.7	**2.5 – 6.3**	**2.9 – 6.0**	**1.0 – 1.2**
Meas.	**0.74**	**1.44**	**2.56**	2.91	**4.77**	**4.27**	**1.36**
Meas.SD	**0.42**	**0.54**	**0.68**	0.85	**1.11**	**1.69**	**0.30**

Pesticides (N=4)

	pp'-DDT*	**pp'-DDD**	**pp'-DDE**	Lindan*	Diazinon*	**Heptachlor**	Heptaepx.A*	Heptaepx.B*	Endosulf I*	Endosulf II*	**Aldrin**	Dieldrin*	Endrin*
Cert.	*3.0*	***2.8***	***9.2***	0.27	na	**0.32**	0.99		3.5	1.4	***0.75***	3.5	1.9
Cert. 95% CI	*1.8 – 5.6*	***2.0 – 3.7***	***6.2 - 11***	0.11 – 0.80	na	**0.23 – 0.46**	0.37 – 1.6		0.94 – 4.7	1.0 – 1.6	***0.61 – 1.2***	1.4 – 7.0	0.86 – 5.1
Meas.	3.58	**2.68**	**6.15**	0.64	nd	**0.45**	0.47	0.65	3.04	0.94	**0.81**	4.92	5.30
Meas.SD	0.50	**0.74**	**1.78**	0.37	na	**0.18**	0.14	0.17	0.86	0.48	**0.20**	1.30	0.67

* information values
na = not available
nd = not detected

Table S3. Individual total length, calculated age, weight, and muscle tissues physiological parameters and isotopic $\delta^{15}N$ and $\delta^{13}C$ values measured for the 23 European conger specimens caught in the Gulf of Fos.

Site	Length (cm)	Estimated age (y)	Weight (g)	Lipid (% ww)	Water (%)	$\delta^{15}N$ (‰)	$\delta^{13}C$ (‰)
l	75	4.3	800	0.27	68.9	12.18	-17.13
m	78	4.6	700	0.33	70.8	13.39	-16.82
k	78	4.6	710	0.29	73.2	12.68	-16.44
n	78	4.6	900	0.39	70.4	13.14	-17.44
g	87	5.5	1100	0.36	74.6	14.02	-16.44
f	93	6.1	1300	0.26	74.2	13.89	-17.62
f	94	6.2	1300	0.36	72.6	13.17	-17.05
f	96	6.4	1600	0.32	74.7	13.92	-17.63
f	100	6.9	2400	0.38	75.0	14.29	-17.34
f	108	7.8	2500	0.22	74.5	14.50	-16.56
e	108	7.8	2200	0.06	78.4	13.80	-16.04
c	116	8.7	3000	0.54	75.8	13.36	-16.93
h	117	8.9	3300	0.17	76.8	14.10	-16.98
d	120	9.2	3190	0.22	76.2	13.84	-15.97
c	122	9.5	3700	0.99	72.6	13.87	-16.93
g	125	9.9	3500	0.10	77.9	14.07	-16.93
a	126	10.0	4250	0.87	74.4	13.22	-16.79
j	127	10.2	3400	0.10	78.0	14.68	-15.45
a	133	11.0	4550	0.58	76.0	13.54	-16.79
i	133	11.0	4900	1.21	74.3	15.33	-15.01
b	135	11.3	5140	0.24	76.4	14.25	-16.21
e	137	11.5	5600	0.83	74.6	13.80	-16.20
i	140	12.0	5700	0.71	76.9	14.02	-16.18

Table S4. Individual metal concentrations (mg.kg^{-1} ww) measured for the 23 European conger specimens caught in the Gulf of Fos (Co, Ag, Cd, Sn, Sb, Tl, Pb not detected).

Site	length	Al	*Cr*	Mn	Fe	Ni	Cu	As	Mo	Hg	Zn
l	75	<0.03	0.28	0.10	3.64	0.19	2.89	11.82	0.06	0.35	2.02
m	78	0.33	2.17	0.37	9.92	0.85	0.54	13.43	<0.01	<0.01	3.21
k	78	0.21	0.72	0.32	3.76	0.11	0.45	18.52	<0.01	0.41	2.95
n	78	0.23	1.62	0.42	5.92	0.38	0.53	6.52	<0.01	<0.01	2.37
g	87	<0.03	1.04	0.21	2.75	0.64	2.75	10.69	0.11	0.35	2.70
f	93	<0.03	0.34	0.16	1.37	0.16	2.38	5.94	0.05	0.23	1.34
f	94	0.22	1.89	0.35	7.40	0.65	0.50	7.13	<0.01	0.17	1.92
f	96	0.21	0.98	0.35	7.60	0.18	0.43	8.11	<0.01	1.35	3.29
f	100	0.07	0.46	0.17	2.80	0.07	1.78	11.76	0.04	0.26	2.85
f	108	0.05	0.13	0.08	0.79	0.02	1.12	21.71	0.04	0.14	2.09
e	108	<0.03	0.14	0.06	0.78	0.05	0.80	5.19	0.03	0.12	1.12
c	116	0.10	0.58	0.17	2.11	0.12	4.21	12.84	0.05	0.22	2.30
h	117	0.12	1.16	0.37	6.09	0.26	6.76	21.15	0.09	0.35	4.32
d	120	0.24	0.38	0.13	1.88	0.06	4.59	27.12	0.05	0.56	4.04
c	122	<0.03	0.52	0.06	2.49	0.18	0.82	9.85	0.09	0.19	2.76
g	125	0.04	0.21	0.06	1.17	0.08	1.19	8.85	0.04	0.21	1.37
a	126	<0.03	0.19	0.08	0.90	0.01	2.31	55.61	0.03	0.53	2.66
j	127	0.05	0.85	0.47	3.38	0.23	2.53	17.35	0.05	0.19	2.68
a	133	0.09	0.48	0.27	2.18	0.08	5.25	60.14	0.03	0.73	7.43
i	133	0.06	0.70	0.43	2.81	0.25	4.74	16.73	0.05	0.27	5.41
b	135	0.14	0.76	0.31	7.11	0.17	7.09	42.76	0.05	0.61	6.19
e	137	0.06	0.51	0.07	9.75	0.12	1.53	12.47	0.07	0.28	5.17
i	140	0.03	0.08	0.06	0.53	<0.01	1.69	10.16	0.03	0.16	2.29

Table S5. Individual PAH concentrations (µg.kg^{-1} ww) measured for the 23 European conger specimens caught in the Gulf of Fos.

Site	length	Nap	Acy	Ace	Flu	Phe	Ant	FlA	Pyr	BaA	Chr	BbF	BkF	BaP	DBA	BPe	Ipy	TOTAL
l	75	0.96	0.27	0.69	0.17	0.61	0.24	0.12	0.17	0.09	0.12	0.08	0.07	0.08	0.07	0.05	1.11	4.91
m	78	1.44	0.25	1.06	0.25	0.59	0.22	0.19	0.24	0.12	0.15	0.12	0.08	0.08	0.07	0.06	1.25	6.16
k	78	0.96	0.22	0.63	0.21	0.64	0.25	0.14	0.17	0.09	0.11	0.07	0.05	0.05	0.06	0.04	1.09	4.79
n	78	0.89	0.27	0.61	0.26	0.97	0.24	0.16	0.23	0.11	0.10	0.09	0.07	0.09	0.07	0.04	1.04	5.24
g	87	0.92	0.21	0.48	0.22	0.65	0.21	0.18	0.22	0.12	0.14	0.13	0.09	0.09	0.06	0.05	0.87	4.66
f	93	0.69	0.23	0.44	0.20	0.52	0.13	0.08	0.12	0.07	0.09	0.07	0.05	0.05	0.05	0.04	0.79	3.64
f	94	0.90	0.23	0.55	0.24	0.55	0.27	0.23	0.16	0.09	0.18	0.10	0.07	0.07	0.06	0.05	0.92	4.66
f	96	0.63	0.20	0.46	0.21	0.43	0.24	0.13	0.14	0.08	0.08	0.06	0.05	0.06	0.05	0.04	0.81	3.67
f	100	1.48	0.27	1.49	0.64	0.66	0.34	0.31	0.40	0.12	0.18	0.16	0.26	0.25	0.08	0.09	0.81	7.55
f	108	2.05	0.36	1.09	0.34	0.63	0.28	0.23	0.30	0.20	0.28	0.15	0.11	0.13	0.22	0.16	1.21	7.75
e	108	1.11	0.18	1.00	0.22	0.45	0.70	0.23	0.29	0.16	0.17	0.15	0.22	0.22	0.13	0.08	0.88	6.19
c	116	1.38	0.34	0.78	0.50	0.85	0.25	0.30	0.37	0.12	0.11	0.16	0.25	0.23	0.08	0.07	0.83	6.62
h	117	1.14	0.29	0.83	0.48	0.70	0.43	0.43	0.51	0.21	0.23	0.29	0.30	0.26	0.08	0.12	0.85	7.14
d	120	1.25	0.25	0.93	0.39	0.71	0.21	0.23	0.35	0.14	0.14	0.18	0.26	0.25	0.08	0.09	0.96	6.41
c	122	1.34	0.71	1.04	0.63	0.64	0.85	0.37	0.24	0.15	0.29	0.14	0.26	0.22	0.16	0.11	1.06	8.20
g	125	1.37	0.41	0.72	0.50	0.57	0.56	0.28	0.33	0.14	0.20	0.16	0.26	0.22	0.14	0.11	0.88	6.83
a	126	2.39	0.68	0.69	0.91	0.70	1.37	0.25	0.32	0.16	0.18	0.14	0.24	0.21	0.15	0.11	0.97	9.48
j	127	1.39	0.41	0.95	0.19	0.38	0.30	0.40	0.26	0.20	0.25	0.15	0.11	0.14	0.19	0.15	0.95	6.43
a	133	1.26	0.49	1.03	0.59	0.58	0.28	0.23	0.27	0.13	0.15	0.12	0.23	0.20	0.07	0.07	1.05	6.74
i	133	1.60	0.51	1.09	0.44	1.09	0.28	0.39	0.43	0.24	0.58	0.22	0.15	0.20	0.15	0.16	0.97	8.49
b	135	1.36	0.45	1.38	0.50	0.52	0.22	0.31	0.39	0.13	0.21	0.16	0.19	0.23	0.07	0.05	0.96	7.13
e	137	1.29	0.29	0.97	0.56	0.66	0.82	0.28	0.24	0.20	0.30	0.17	0.26	0.24	0.13	0.14	1.06	7.61
i	140	1.48	0.82	0.86	0.79	0.83	0.23	0.35	0.38	0.18	0.24	0.18	0.25	0.24	0.14	0.12	0.83	7.90

Table S6 (1). Individual PCB concentrations (µg.kg^{-1} ww) measured for the 23 European conger specimens caught in the Gulf of Fos (nd = not detected).

Site	length	PCBi	PCB-DL	CB 18	CB 20	CB 28	CB 31	CB 44	CB 52	CB 84	CB 87	CB 92	CB 95	CB 99	CB 101	CB 105	CB 118	CB 128
l	75	20.23	1.76	0.14	0.00	0.03	0.17	0.19	0.13	0.06	0.16	0.05	0.40	0.12	0.75	0.13	1.57	0.41
m	78	9.61	2.32	0.00	0.00	0.09	0.08	0.33	0.26	0.14	0.48	0.11	0.73	0.19	1.34	0.22	2.05	0.32
k	78	14.06	3.32	0.25	0.11	0.06	0.37	0.34	0.65	0.37	0.66	0.20	0.77	0.37	1.38	0.40	2.87	0.49
n	78	16.33	3.20	0.07	0.28	0.16	0.26	0.67	1.18	1.84	1.12	0.69	0.79	1.53	1.41	0.56	2.27	0.41
g	87	9.09	2.73	0.28	0.14	0.07	0.10	0.34	0.96	0.12	0.36	0.06	0.66	0.16	0.97	0.15	2.49	0.31
f	93	9.14	1.62	0.13	0.14	0.03	0.25	0.05	0.18	0.05	0.19	0.06	0.44	0.12	0.83	0.13	1.29	0.32
f	94	11.90	1.37	0.15	0.00	0.04	0.07	0.43	0.26	0.08	0.18	0.07	0.47	0.10	0.68	0.06	1.26	0.18
f	96	7.15	1.33	0.14	0.14	0.04	0.31	0.43	0.22	0.06	0.18	0.06	0.50	0.14	0.86	0.11	1.10	0.24
f	100	12.91	0.84	nd	0.00	0.16	0.24	0.05	0.33	0.05	0.22	0.08	0.47	0.19	1.10	0.08	0.73	0.31
f	108	8.99	1.36	0.04	nd	0.08	0.09	0.05	0.15	0.03	0.08	0.13	0.17	0.27	0.20	0.15	1.17	0.19
e	108	7.55	0.68	nd	0.00	0.13	0.00	0.02	0.10	0.02	0.07	0.03	0.16	0.08	0.47	0.11	0.54	0.15
c	116	18.06	1.61	0.09	0.02	0.14	0.20	0.07	0.25	0.05	0.25	0.14	0.61	0.33	1.69	0.21	1.39	0.44
h	117	13.35	0.83	nd	nd	0.14	0.21	0.09	0.22	0.04	0.16	0.09	0.37	0.20	0.91	0.05	0.72	0.32
d	120	8.43	0.98	0.07	nd	0.17	0.23	0.03	0.30	0.05	0.16	0.03	0.28	0.09	0.63	0.16	0.80	0.18
c	122	28.67	2.08	0.03	0.01	0.16	0.00	0.22	0.18	0.02	0.21	0.26	0.76	0.51	2.35	0.13	1.80	0.63
g	125	9.03	0.71	0.07	nd	0.17	0.06	0.03	0.18	0.02	0.09	0.05	0.24	0.10	0.61	0.02	0.66	0.20
a	126	22.55	0.85	0.06	nd	0.13	0.17	0.06	0.18	0.02	0.15	0.12	0.43	0.26	1.29	0.05	0.75	0.40
j	127	9.60	0.72	nd	nd	0.08	nd	0.05	0.12	0.01	0.08	0.03	0.17	0.10	0.49	0.06	0.64	0.23
a	133	16.44	1.04	nd	nd	0.17	0.24	0.05	0.44	0.05	0.22	0.08	0.45	0.21	0.88	0.09	0.93	0.38
i	133	19.17	1.39	0.01	0.07	0.17	0.23	0.09	0.22	0.02	0.13	0.13	0.31	0.32	0.90	0.15	1.22	0.47
b	135	11.06	1.89	0.06	nd	0.16	0.20	0.02	0.26	0.07	0.24	0.08	0.42	0.16	0.63	0.13	1.70	0.28
e	137	21.13	1.86	0.01	0.03	0.15	0.22	0.03	0.14	0.03	0.15	0.13	0.43	0.30	1.30	0.20	1.62	0.47
i	140	21.82	1.43	0.05	0.07	0.17	0.21	0.06	0.16	0.03	0.18	0.19	0.62	0.35	1.51	0.26	1.14	0.56

Table S6 (2). Individual PCB concentrations (µg.kg^{-1} ww) measured for the 23 European conger specimens caught in the Gulf of Fos.

Site	length	CB 135	CB 136	CB 138	CB 141	CB 149	CB 151	CB 153	CB 156	CB 170	CB 174	CB 177	CB 180	CB 183	CB 187	CB 194	CB 195	CB 196	CB 201
l	75	0.08	0.03	4.31	0.36	1.26	0.58	8.86	0.06	1.80	0.39	1.03	6.14	1.49	4.40	1.24	0.35	1.16	1.44
m	78	0.07	0.06	2.43	0.25	1.10	0.36	3.35	0.05	0.52	0.27	0.45	2.14	0.46	1.48	0.38	0.09	0.33	0.40
k	78	0.09	0.09	3.49	0.41	1.25	0.27	5.51	0.05	0.77	0.24	0.48	2.97	0.67	2.14	0.53	0.14	0.41	0.59
n	78	0.07	0.21	3.72	0.44	1.75	0.31	6.40	0.37	0.76	0.07	0.37	3.46	0.83	2.26	0.79	0.50	0.52	0.43
g	87	0.05	0.08	2.05	0.20	1.66	0.17	3.28	0.10	0.37	0.30	0.57	1.76	0.46	1.63	0.34	0.10	0.53	0.72
f	93	0.06	0.04	2.20	0.20	0.97	0.34	3.84	0.20	0.48	0.27	0.42	2.06	0.53	1.58	0.34	0.11	0.34	0.34
f	94	0.05	0.03	2.04	0.15	0.92	0.30	4.60	0.04	0.98	0.22	0.54	4.28	0.76	2.66	1.11	0.23	0.99	1.01
f	96	0.05	0.04	1.78	0.15	0.78	0.33	2.84	0.11	0.35	0.16	0.46	1.41	0.36	1.16	0.28	0.05	0.44	0.32
f	100	0.07	0.09	3.40	0.25	0.62	0.34	5.33	0.03	0.72	0.31	0.64	2.59	0.70	1.94	0.43	0.12	0.49	0.43
f	108	0.05	0.05	2.07	0.17	1.05	0.34	4.25	0.04	0.68	0.25	0.48	2.25	0.57	1.78	0.42	0.11	0.53	0.49
e	108	0.03	0.02	1.68	0.14	0.35	0.20	3.43	0.04	0.62	0.13	0.44	1.73	0.44	1.38	0.30	0.09	0.34	0.36
c	116	0.07	0.09	4.48	0.33	0.97	0.22	8.46	0.02	1.01	0.44	0.80	3.06	0.97	3.24	0.62	0.14	0.58	0.63
h	117	0.10	0.06	3.42	0.31	0.72	0.69	5.83	0.06	0.98	0.46	0.75	2.84	0.86	2.35	0.54	0.16	0.61	0.55
d	120	0.02	0.02	1.82	0.16	0.75	0.05	3.40	0.02	0.72	0.13	0.49	2.12	0.47	1.55	0.38	0.11	0.40	0.42
c	122	0.18	0.07	6.66	0.51	1.53	1.26	14.1	0.15	1.96	0.69	1.45	5.23	1.61	5.88	1.17	0.38	1.15	1.30
g	125	0.04	0.02	2.10	0.16	0.81	0.39	4.00	0.03	0.62	0.25	0.52	1.97	0.57	1.70	0.38	0.11	0.39	0.43
a	126	0.09	0.04	4.94	0.33	0.77	0.55	10.9	0.04	1.39	0.40	0.90	5.10	1.28	3.70	0.86	0.29	0.85	0.88
j	127	0.03	0.02	2.33	0.16	0.49	0.22	4.56	0.02	0.56	0.17	0.61	2.02	0.44	1.99	0.37	0.12	0.26	0.39
a	133	0.04	0.06	4.01	0.27	1.16	0.15	7.52	0.02	1.01	0.32	0.75	3.43	0.90	2.50	0.66	0.20	0.61	0.60
i	133	0.07	0.03	5.73	0.28	0.74	0.37	9.37	0.03	1.03	0.31	0.88	2.78	0.95	2.84	0.47	0.17	0.43	0.49
b	135	0.04	0.07	2.85	0.22	1.60	0.10	5.00	0.07	0.75	0.24	0.58	2.16	0.62	2.00	0.37	0.11	0.46	0.46
e	137	0.11	0.04	4.89	0.34	1.61	0.71	10.1	0.05	1.64	0.41	1.04	4.53	1.20	3.62	0.88	0.25	0.79	0.86
i	140	0.16	0.06	5.41	0.45	1.72	1.12	10.2	0.02	1.75	0.66	1.05	4.40	1.36	3.87	0.86	0.24	0.89	0.82

Table S7. Individual pesticide concentrations (µg.kg^{-1} ww) measured for the 23 European conger specimens caught in the Gulf of Fos.

Site	**length**	lindan	Diaz-inon	Hepta chlor	Hepta epx.A	Hepta epx.B	Endos ulfan I	Endos ulfanII	aldrin	diel-drin	endrin	**total**
l	75	0.12	0.36	0.04	0.03	0.08	0.31	0.91	0.00	0.53	0.01	***2.40***
m	78	0.19	0.57	0.01	0.05	0.22	0.68	0.63	0.01	0.11	0.03	***2.50***
k	78	0.48	0.44	0.01	0.13	0.53	1.34	0.80	0.01	0.31	0.03	***4.07***
n	78	0.24	0.67	4.24	1.40	4.16	5.06	0.95	0.03	1.29	0.38	***18.42***
g	87	0.26	0.85	0.10	0.07	0.16	0.09	0.61	0.01	0.01	0.02	***2.17***
f	93	0.19	0.64	0.06	0.07	0.16	0.46	0.75	0.00	0.01	0.02	***2.36***
f	94	0.32	0.48	0.01	0.03	0.14	0.01	0.89	0.01	0.56	0.02	***2.48***
f	96	0.59	0.16	0.05	0.04	0.14	0.43	0.47	0.02	0.01	0.01	***1.91***
f	100	0.24	0.50	0.41	0.19	0.15	1.16	1.65	0.29	0.09	0.25	***4.93***
f	108	0.12	0.53	0.09	0.05	0.10	0.22	1.34	0.04	0.92	0.06	***3.47***
e	108	0.28	0.52	0.61	0.20	0.19	1.05	1.61	0.23	0.76	0.15	***5.59***
c	116	0.28	0.27	0.54	0.26	0.25	1.80	2.38	0.30	0.15	0.42	***6.65***
h	117	0.19	0.40	0.26	0.19	0.12	0.55	1.69	0.35	0.03	0.03	***3.82***
d	120	0.27	0.58	0.22	0.13	0.24	0.30	1.52	0.01	0.03	0.10	***3.41***
c	122	0.19	0.43	0.47	0.26	0.12	1.78	3.86	0.25	0.08	0.12	***7.56***
g	125	0.28	0.54	0.10	0.09	0.13	0.04	1.51	0.04	1.54	0.08	***4.35***
a	126	0.14	1.24	0.08	0.13	0.18	0.02	2.24	0.02	0.04	0.04	***4.14***
j	127	0.13	0.57	0.08	0.06	0.12	0.46	1.71	0.03	0.48	0.04	***3.69***
a	133	0.28	0.63	0.32	0.25	0.17	1.31	2.15	0.35	0.09	0.20	***5.75***
i	133	0.15	0.65	0.16	0.15	0.15	0.03	2.32	0.03	0.04	0.14	***3.81***
b	135	0.31	0.60	0.52	0.19	0.36	0.76	1.78	0.29	0.03	0.09	***4.94***
e	137	0.27	1.17	0.63	0.15	0.13	1.45	2.96	0.28	0.30	0.11	***7.45***
i	140	0.12	0.77	0.10	0.18	0.16	0.01	2.91	0.02	0.05	0.06	***4.38***

Table S8. Tolerable daily intakes (TDI) values employed, for chronic oral exposure. Most of the TDI values were given by the French National Institute for Industrial Environment and Risks latest TDI compilation (INERIS, 2009 and 2015), prepared for the French government, and by the JECFA database. When not available, it was completed by the WHO, EFSA, or US agencies data.

	TDI ($\mu g.kg^{-1}.d^{-1}$)	source value type	source
Al	143	PTDI	3
As inorganic	0,3	RfD/MRL	1
Cr (total)	300	TDI	6
Cu	140	MRL	1
Fe	800	PMTDI	3
Hg total	0,57	PTDI	3
Mn	140	RfD	10
Mo	4100	TDI	4
Ni	2,8	TDI	7
Zn	300	PMTDI	3
benzo(a)pyrene	0,005	DVS	1
PCBi	0,02	MRL	1
PCB-DL	$0,7.10^{-6}$ (ITEQ)	PTDI	2
aldrin	0,03	RfL	8
dieldrin	0,05	RfL	8
endrin	0,2	TDI	5
diazinon	0,2	TDI	5
endosulfan I+II	6	TDI	5
Heptachlor and epoxydes	0,1	TDI	5
lindan	0,01	MRL	9

[1] INERIS: Overview of reference toxicological values (2009, in French)

[2] INERIS: Overview of reference toxicological values, compilation table update (2015, in French)

[3] JECFA: Evaluations of the Joint FAO/WHO Expert Committee on Food Additives (JECFA), World Health Organization (last accessed in December 2018).

[4] WHO: Molybdenum in drinking water. Background document for development of WHO guidelines for drinking water quality (2011)

[5] EFSA: The 2013 European Union report on pesticide residues in food., EFSA Journal 13 : 4038 (2015).

[6] EFSA CONTAM Panel: Scientific opinion on the risks to public health related to the presence of chromium in food and drinking water (2014)

[7] EFSA CONTAM Panel: Scientific opinion on the risks to public health related to the presence of nickel in food and drinking water (2015)

[8] USEPA: Integrated Risk Information System (IRIS). Chemical assessment summaries. National Center for Environmental Assessment, U.S. Environmental Protection Agency (last updated in December 2018).

[9] ATDSR: Minimal Risk Levels (MRLs) List, Minimal risk levels (MRLs) for hazardous substances, Agency for Toxic Substances & Disease Registry (last updated in August 2018).

[10] National Food Agency, Sweden (Livsmedelsverket). Contaminants and minerals in foods for infants and young children, part 2: risk and benefit assessment (2013)