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Organochlorine residues in muscle of European eels (*Anguilla anguilla*) from four Spanish Mediterranean wetlands and coastal lagoons

Alonso Pérez-Vegas [a], Marcos Pérez-López [b], Elena Barcala [d], Diego Romero [c,*], Pilar Muñoz [a]

[a] Department of Animal Health, Regional Campus of International Excellence "Campus Mare Nostrum", Universidad de Murcia, 30100 Murcia, Spain
[b] Toxicology Unit, Veterinary School, Avda. de la Universidad s/n, 10003 Cáceres, Spain
[c] Toxicology Department, Regional Campus of International Excellence "Campus Mare Nostrum", Universidad de Murcia, 30100 Murcia, Spain
[d] Centro Nacional Instituto Español de Oceanografía, Centro Oceanográfico de Murcia, CSIC, C/Varadero, s/n, 30740 San Pedro del Pinatar, Spain

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ABSTRACT

European eels (*Anguilla anguilla*) are an endangered species throughout their range, and chlorine organic compounds are some of the most important pollutants for marine species. Data on contaminants in eel stocks remain incomplete, so organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in muscle of European eels from four Spanish Mediterranean ecosystems were analyzed. COPs are presents in eels from all areas, but some compounds are not detected: HCH α, β and γ (lindane), endosulfan sulfate, heptachlor, and PCBs 28, 52 and 180. The high percentage of DDT 2,4′ in eels from S'Albufera des Grau Natural Park, an ecosystem with good ecological status according to the Water Framework Directive, and the presence of PCBs in S'Albufereta Natural Reserve indicate the need to carry out further studies in the future. The results obtained can improve the management of this species in the studied areas.

1. Introduction

Human activities are well-known sources of contaminants that can seriously damage the environment, a good example being oceanic contamination and the effects it has on wildlife. Chlorine organic pollutants (COPs), as organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) are some of the most serious contaminants of natural systems due to their frequency of use over past decades and the fact that a non-negligible proportion of their active compounds does not reach the targeted organisms (Carles et al., 2021). Even though their use has been banned in many countries by the Stockholm Convention (UNEP, 2018), they are still present in many marine environments (e.g. Neves et al., 2017; Olisah et al., 2021). They end up in rivers, seas and oceans, and can be absorbed by a wide range of organisms (e.g. Windsor et al., 2019). Apart from absorption through gills and skin, carnivorous fish are particularly vulnerable to high contamination levels due to the effects of bioaccumulation and biomagnification (Belpaire and Goemans, 2007).

Environmental pollutants as COPs are considered endocrine-disrupting by their ability to bind with estrogen receptors (see review in Mills and Chichester, 2005). OCPs have been proven to interfere with several corporal functions and metabolic systems in fauna. They disturb the normal function of the thyroid hormonal system (Brown et al., 2004; Langer, 2010), act as anti-androgens (Kelce et al., 1997) and affect the immune system (Martyniuk et al., 2016a, 2016b). OCP bioaccumulation can also impair reproductive migration in certain fish species since the exposure to OCPs decreases speed, swimming distances and body-turn angles (Pereira et al., 2012); furthermore, it also causes a highly significant decrease in testicular testosterone concentrations during the spawning season and lowers spermatogonia number and size (Islam et al., 2017). Toxicological studies confirmed that PCBs can cause effects on the immune system (Henry, 2015); they are stressors to fish by metabolic disturbance and alteration of swimming performance (Bellehumeur et al., 2016), they can also alter liver ultrastructure, and reduce the fecundity and hatching rate (Hugla and Thomé, 1999). The effects of COPs are particularly relevant in the European eel (*Anguilla anguilla*) (Geeraerts and Belpaire, 2010; van Ginneken et al., 2009a), a species listed as Critically Endangered on the International Union for Conservation of Nature Red List (Pike et al., 2020), and the object of European Eel Regulation EC No. 1100/2007 (European Commission, 2007) implemented via a number of Eel Management Plans. This panmictic catadromous species conducts an extraordinary 5000–7000-km journey to the Sargasso Sea in the North Atlantic Ocean to spawn, from where

* Corresponding author at: Área de Toxicología, Facultad de Veterinaria, Campus Regional de Excelencia Internacional Campus Mare Nostrum, Universidad de Murcia, 30100 Murcia, Spain.
E-mail address: diegorom@um.es (D. Romero).

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their larvae travel all the way back to Europe (Schmidt, 1923; Miller et al., 2015).

Causative relationships between COP exposure and its biological effects are difficult to demonstrate. Nevertheless, effects in European eels have been reported at several levels of biological organization, from subcellular to organ, individual and population levels (ICES, 2018). For instance, Palstra et al. (2006) artificially stimulated female and male silver eels to reach maturation and breed, and then studied the effects of dioxin-like compounds on muscle and gonad tissues during embryonic development. They reported great differences in the embryonic development of eel eggs and a correlation between embryo survival time and PCBs levels in the gonads. These disrupting effects occurred at levels below the EU eel-consumption standard. van Ginneken et al. (2009b) used swimming tunnels to demonstrate that PCB-exposure significantly reduces oxygen consumption during swimming, an effect that begins to have an impact at 400 km and then increases as time passes.

The EIFAC/ICES Working Group on Eels (WGEEL) set up in September 2007 a European Eel Quality Database to collate recent data on contaminants and diseases throughout the distribution area of the European eel (Belpaire et al., 2011a). However, data on contaminants in eel stocks remain incomplete and so efforts are needed to update and extend this database (ICES, 2021). To improve insight into the current status of pollution by COPs in Mediterranean European eel populations, our study examined eels from four different Spanish locations affected by varying degrees of anthropogenic impact. Spatial variation in the level of COP pollution could shed light on the areas of most concern for contamination by these substances. Our initial hypothesis was that COPs can be present in eels from all studied ecosystems, so the reporting of the levels found could be useful in the population management.

2. Material and methods

2.1. Sampling area

Eels were obtained from four Spanish Mediterranean ecosystems subjected to different degrees of anthropogenic pressure: i) S'Albufera des Grau Natural Park (AG), ii) S'Albufereta Natural Reserve (ANR), iii) El Hondo-Salinas de Santa Pola Natural Park (EH-SP), and iv) Albufera de Valencia (AV) (Fig. 1). These ecosystems are of great ecological value since they are Special Protection Areas for Birds, Sites of Community

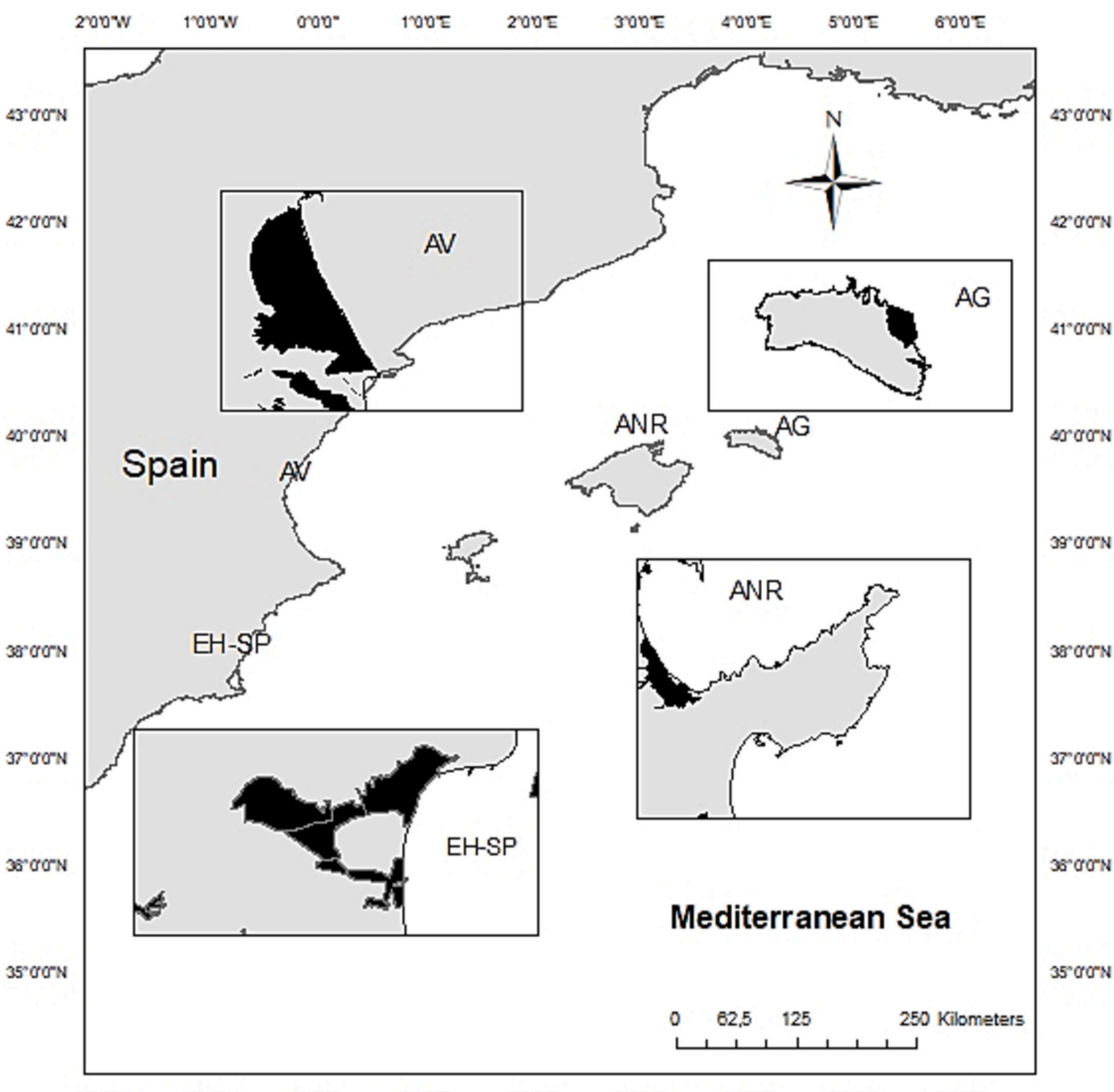

Fig. 1. Location of sampling areas from **Spanish Mediterranean wetlands and coastal lagoons.**

Interest (AG, ANR, AV), Natura 2000 areas (AG, ANR, AV) and important wetlands included in the RAMSAR convention (ANR, EH-SP, AV).

S'Albufera des Grau Natural Park (39°58′35″N 4°14′23″E) on the Balearic Island of Menorca has a surface area of 78 ha, with an average depth of 1.37 m and a maximum depth of 3 m. The lagoon receives freshwater from two streams that drain an area of 56 km^2. These freshwater inputs are irregular and occur largely in spring and autumn. The lagoon is connected to the sea by a narrow, 500-m-long channel, Sa Gola, in which a small floodgate allows the lagoon-sea connection to be regulated when the sand barrier is open. The water of the lagoon is mesohaline with a salinity of 6–20 PSU. There is marked seasonality in the salinity and water levels due to the Mediterranean evaporation/precipitation regime. According to the Water Framework Directive, the ecological status of this site is good to very good, the main threats being the spillage of slurry and the fall in the amount of inflowing freshwater (Govern de les Illes Balears, 2015).

S'Albufereta Natural Reserve (39°86′305″N 03°088′60″E) is located in the bay of Pollença, in the northeast of the island of Mallorca (Balearic Islands). It is one of the most important wetlands in Mallorca, with 211,43 ha protected as a Special Natural Reserve, a buffer zone of 301,82 ha, and a 100-m security strip around the strict protection zone. This wetland lies the confluence of four freshwater streams, the largest of which is Torrent del Rec. These streams run down to a plain next to the sea bordered by a dune chain, open to the sea through Es Grau, and by a peripheral fifth stream. It enjoys a Mediterranean climate, with periods of summer drought and an average annual rainfall of about 708 mm. It is separated from the sea to the east by a 100-m-wide sand bar, while inland it is surrounded by semi-abandoned farmland. According to the Water Framework Directive, its ecological status is good, the main threats to its conservation being spillage from sewage treatment and infiltration from septic tanks (Govern de les Illes Balears, 2015).

El Hondo-Salinas de Santa Pola Natural Park (38°11′N, 00°45′W) lies a rural environment in the south of Alicante province, where two natural parks are crisscrossed and interconnected by an extensive network of canals and ponds. El Hondo consists of a complex of mesosaline and polysaline semi-artificial wetlands used to store irrigation water, while the neighbouring saltpans of Salinas de Santa Pola are fed by inflow from agricultural areas. These ecosystems are regarded as one of the most important wetlands in south-east Spain given that they are of vital importance for a vast variety of fauna and flora including many migratory birds. The main threats to these sites are low water levels and poor water quality (CHS, 2020).

Albufera de Valencia (39°19′54″N 0°21′08″O) is a shallow hypertrophic lagoon (salinity 1–2 ‰, depth 1–3 m), situated 15 km south of the city of Valencia, with a surface area of 23.2 km^2 (the second largest coastal lagoon in Spain). It is a freshwater lagoon since its connection to the sea is regulated and, according to the quality variables used by the Water Framework Directive (CHJ-GV-AV, 2019), it has a poor ecological status. Its poor trophic state is due to pressure from surrounding urban and agricultural environments that contaminate inflowing water with nutrients and phytosanitary products (CHJ-GV-AV, 2019).

2.2. Sampling

Eels were either bought from local fishermen in AV (n = 30) and EH-SP (n = 25) or fished using traditional gear in EH-SP (n = 5), ANR (n = 28) and AG (n = 30). Eels were sacrificed by sedation followed by an overdose of tricaine methanesulfonate (MS222) at 100 mg/L in accordance with current legislation. The total length to the nearest millimeter and the total weight to the nearest gram were measured. Eels were then dissected to obtain sections (1–1.5 g) of muscle from an area 4–5 cm behind the anal cavity, which were then stored at −20 °C until processed.

2.3. Quantification of chlorinated compounds and GC/MS analysis

A total of 20 OCPs were assayed: isomer mixture of hexachlorocyclohexane (HCH) consisting of α, β and γ-HCH; 2,4′- and 4,4′-DDT and its metabolites (namely 2,4′- and 4,4′-DDD, as well as 2,4′- and 4,4′-DDE); chlordane (cis and trans); and the cyclodiene insecticides endosulfan (I and II) and endosulfan sulfate, heptachlor and heptachlor epoxide A and B, aldrin, endrin and dieldrin. Similarly, seven indicator PCBs (CBs 28, 52, 101, 118, 138, 153 and 180) were targeted as they are often present in biotic and abiotic matrices, and are recognized as representative of the whole group of PCBs by the Agency for Toxic Substances and Disease Registry (ATSDR, 2020).

Reference materials with a purity of 97–99.7 % (Dr. Ehrenstorfer GmbH, Augsburg, Germany) were used for the standard OCP preparation (final concentrations ranging from 10 ppb to 10 ppm). Likewise, a commercial mix of seven PCBs (SpexCertiPrep, Stanmore, UK) (10 μg/mL in isooctane) was used for the single quantification of PCB congeners IUPAC 28, 52, 101, 118, 138, 153 and 180. Stock solutions (500 μg/mL) were prepared daily by dissolving the reference standards in acetone (Panreac). Working solutions for sample fortification and for injection in analytical systems were prepared by diluting stock solutions in n-hexane (Panreac®).

The extraction protocol used in this study was adapted from a procedure employed by Mateo et al. (2012): samples were thawed at room temperature and 0.7 g of the tissue was chopped and mixed with 7 mL of n-hexane. The mixture was then homogenized and frozen overnight to allow the fat to precipitate. Five mL of the supernatant were added to 2 mL of H_2SO_4; the tubes were subsequently shaken in an orbital shaker for 10 min, sonicated for 5 min and centrifuged at 1000 ×*g* for 5 min, and the acid-containing phase discarded. The above procedure was repeated until the acidic phase was completely clear. The obtained extract was subsequently evaporated, re-suspended in 200 μL n-hexane and used for the OCP and PCB concentration measurements.

A Bruker Scion 456 triple quadrupole gas chromatograph mass spectrometer was used to analyze the samples. Analyte separation was performed using a Rxi-5 Sil MS column (30 m × 0.25 mm, i.d. × 0.25 film thickness). The results were analyzed using specific GCMS software. The multiple-ramp temperature program used involved a first step of 3.5 min at 70 °C, then the temperature was raised to 180 °C at a rate of 25 °C/min. This was followed by an increase to 300 °C at a rate of 15 °C/min, and a final increase to 325 °C at a rate of 50 °C/min, which was maintained for 5 min. The vaporized samples were injected in splitless mode at a column flow rate of 1.20 mL/min. The temperatures of the injection port, detector and interface were 280, 280 and 300 °C, respectively. PCB and OCP residues were quantitatively evaluated through the internal standard method (with 25 μg/L of PCB180 added at the beginning of the extraction process). The calibration curves were obtained by determining the relationship between the peak area and the concentrations of the different standards. Solvent blanks (consisting of 500 μL n-hexane instead of tissue) were processed in parallel to the samples to guarantee the quality of the analyses.

To verify the suitability and performance of the procedure, the accuracy was estimated using recovery experiments that analyzed blank muscle tissue samples (n = 10) spiked with five concentration levels of the PCB and OCP mixtures. Previously, the blank samples were analyzed in triplicate to determine their content of analytes. Recoveries were obtained as the ratio (in %) between the calculated concentration of spiked samples and the theoretical added concentrations. The recovery percentages for PCB spiked samples were found to lie between 84 and 104 % (CV < 20 %), while the recovery percentages for OCPs were between 87 and 112 % (CV < 20 %). The limit of quantification (LOQ) was established as the lowest concentration level validated with satisfactory recovery values (70–110 %) and precision (RSD < 20 %). The limit of detection (LOD) was estimated as the analyte concentration that produced a peak signal that was three times the background noise in the chromatogram at the lowest fortification level studied for each compound. The LODs of the analytes present in the blank tissue sample were estimated from the chromatograms corresponding to the analyzed blank sample (Hernández et al., 2005). The LODs for the OCPs and PCBs ranged between 0.070 and 1.124 μg/kg and 0.006 and 0.079 μg/kg lipid weight (l.w.), respectively.

2.4. Statistical analysis

Descriptive data of the COPs were expressed as the number and percentage, mean concentrations (reported on a lipid weight basis, μg/kg l.w.), standard errors and maximums. Due to the non-normal distribution of the data (Kolmogorov–Smirnov for whole population and Shapiro–Wilk test for ecosystem groups), the statistical analyses were performed using non-parametric tests (Kruskal-Wallis and Spearman tests). These statistical tests were limited to those chemicals that could be detected in >35 % of the samples (whole population). Chi-square tests were performed to compare the categorical variables (presence or absence of a compound). For all analyses, p values <0.05 were considered to be significant.

3. Results

Descriptive data of biometric measures (weight and total length) are given in Table 1.

In the whole population, 84.7 % of the eels had <2 COPs, being these percentages between 75 (ANR) and 96.7 % (EH-SP); in addition, 20.3 % of the total number of eels did not have any of the 27 COPs analyzed (26.7 in AV and EH-SP, 17.9 in ANR, and 10.0 % in AG). Of the OCPs, the following compounds were not detected in any of the eel muscle samples: HCH α, β and γ (lindane), endosulfan sulfate and heptachlor. Similarly, PCBs 28, 52 and 180 were not detected. Of the remaining chemical compounds, many were only detected in a very small percentage of samples (<35 %), although given the relevance of the results (especially due to the influence of the sampling area) their main statistical results were considered. Thus, concentrations of OCPs and PCBs in the whole eel population, as well as the number and percentage of samples where the compounds were detected, are shown in Table 2. However, the compounds that were only detected in a single specimen (DDT 4,4′, DDE 2,4′, endosulfan II, heptachlor epoxides A and B, and PCB-153) are not given. The compounds with highest mean concentrations were endosulfan I, DDE 4,4′ and endrin.

The results obtained from the four different sampling areas are shown in Table 3 and Fig. 2. Regarding the number of compounds detected, nine compounds were detected in ANR and AV, and five in AG and EH-SP; DDT 2,4′, DDD 2,4′ and endrin were found in all four areas, and chlordane cis and aldrin in three (Table 3).

The chi-square test shows a statistical association ($p < 0.05$) between the presence of these compounds and eel-catching areas (moderate degree of association, contingency coefficient = 0.39–0.64).

In terms of biometric measures, a highly negative relationship ($p < 0.01$) between DDD 2,4′ concentrations and weight and total length ($r = -0.703$ and $r = -0.715$, respectively), and a low correlation between DDT 2,4′ and total length ($r = -0.302$, $p < 0.05$), were found.

Table 1
Descriptive statistics (mean, standard deviation, and range) of biometric data in European eels from the studied ecosystems: weight (g) and total length (mm).

	Weight	Total length
S'Albufera des Grau Natural Park	259 ± 59 (147–358)	519 ± 74 (195–600)
S'Albufereta Natural Reserve	303 ± 176 (87–776)	575 ± 103 (344–760)
El Hondo-Salinas de Santa Pola Natural Park	801 ± 207 (511–1281)	738 ± 75 (642–915)
Albufera de Valencia	803 ± 135 (597–1098)	743 ± 42 (662–838)

Table 2
Concentrations of OCPs and PCBs (expressed in μg/kg l.w.) in muscle samples of European eels, *Anguilla anguilla* (n = 118): global results. SEM: standard error of mean; LOD: limit of detection; n = number of samples in which the compound was detected.

	Mean ± SEM	Min	Max	n	% above LOD
DDT 2,4′	0.029 ± 0.007	<LOD	0.327	49	41.53
DDD 2,4′	0.269 ± 0.035	<LOD	0.715	21	17.80
DDD 4,4′	0.147 ± 0.028	<LOD	0.221	4	3.39
DDE 4,4′	1.309 ± 0.386	<LOD	2.228	4	3.39
Chlordane cis	0.320 ± 0.118	<LOD	0.893	9	7.63
Chlordane trans	0.043 ± 0.010	<LOD	0.059	3	2.54
Endosulfan I	2.113 ± 1.095	<LOD	3.208	2	1.69
Aldrin	0.563 ± 0.220	<LOD	2.988	14	11.86
Endrin	1.025 ± 0.176	<LOD	4.325	40	33.90
Dieldrin	0.854 ± 0.595	<LOD	2.041	3	2.54
PCB-118	0.604 ± 0.124	<LOD	0.727	2	1.69
PCB-101	0.705 ± 0.153	<LOD	1.427	9	7.63
PCB-138	0.560 ± 0.216	<LOD	1.591	6	5.08

For comparisons between places, the Kruskal-Wallis test was only performed for DDT 2,4′, DDD 2,4′ and endrin. For 2,4′-DDT, the highest mean concentrations were detected in ANR and no statistically significant differences between the other three areas were found; for DDD 2,4′ differences between EH-SP and AG were found; and for endrin, the highest mean concentrations were found in ANR, with significant differences with the other three locations. Fig. 3 shows the concentrations of metabolites for each specimen with DDT 2,4′ > LOD.

4. Discussion

Several long-term monitoring programs show a general decrease in pesticides and PCBs (e.g. Beliaeff et al., 1997), by the reduction in the use and discharge of COPs in the northern hemisphere (Islam and Tanaka, 2004) and by the efficiency of the European risk-management measures (Water Framework Directive in European Communities, 2000, Directive 2000/60/EC, and European Communities, 1976, Directive 76/464/ECC). In the study areas, some studies have reported decreased concentrations of these compounds (Triay, 1995; Deudero et al., 2007; Quijano et al., 2018; Gómez-Ramírez et al., 2019). However, the fluctuations in the concentration in the different organisms (e.g. habitat, climatology, species, feeding behaviour and physiological status) (Carro et al., 2004; Perugini et al., 2004) makes necessary to carry out studies on endangered species.

4.1. Organochlorine pesticides

Although DDT 2,4′ was the dominant organochlorine compound detected in terms of the percentage of positive samples (41.5 %), the highest mean concentrations found were of DDE 4,4′ (1.31 μg/kg, l.w.). In terms of concentrations, the mean residue levels of DDT and its metabolites occurred in the following order: DDE > DDD > DDT, while the ratio DDE/$\sum$DDT (which is used to assess the chronology of DDT inputs) was 0.74. It is interesting to note that a ratio >0.6 suggests an on-going transformation of DDTs in stable ecosystems with no recent inputs of these contaminants (McHugh et al., 2010). The high proportion of DDE found in the analyzed samples compared to DDT is consistent with the long time that has elapsed since the widespread use of this compound was banned in Spain (Bordajandi et al., 2003). DDE was likewise found to be the composite with the highest contributions by Tabouret et al. (2011) and Szlinder-Richert et al. (2010) in French and Polish eels, respectively, as well as by Arai (2014) in Japanese eels. In fact, it is generally assumed that DDE and DDD are more abundant in aquatic ecosystems than DDT (Bonnineau et al., 2016). However, the total percentages of specimens for each DDT and metabolite groups were 42.37 (DDTs), 21.19 (DDDs) and 4.24 (DDEs), while concentrations of metabolites were higher than those found for DDT 2,4′ (Fig. 3); as such,

Table 3
Concentrations of OCPs and PCBs (expressed in µg/kg l.w.) in muscle samples of European eels, *Anguilla anguilla*: results by sampling area. SEM: standard error of mean; LOD: limit of detection; n = number of samples in which the compound was detected.

	S'Albufera des Grau Natural Park (AG)			S'Albufereta Natural Reserve (ANR)			El Hondo-Salinas de Santa Pola (EH-SP)			Albufera de Valencia (AV)		
	Mean ± SEM (range)	n	%	Mean ± SEM (range)	n	%	Mean ± SEM (range)	n	%	Mean ± SEM (range)	n	%
DDT 2,4′	0.023 ± 0.002 (<LOD–0.048)	21	70.00	0.243 ± 0.084 (<LOD–0.327)	2	7.14	0.018 ± 0.002 (<LOD–0.042)	15	50.00	0.017 ± 0.002 (<LOD–0.030)	11	36.67
DDD 2,4′	0.370 ± 0.056 (<LOD–0.715)	10	33.33	0.213 ± 0.005 (<LOD–0.217)	2	7.14	0.173 ± 0.029 (<LOD–0.278)	8	26.67	0.147	1	3.33
DDD 4,4′		0	0.00	0.131 ± 0.027 (<LOD–0.157)	2	7.14		0	0.00	0.163 ± 0.058 (<LOD–0.221)	2	6.67
DDE 4,4′		0	0.00	0.743 ± 0.362 (<LOD–1.105)	2	7.14		0	0.00	1.875 ± 0.354 (<LOD–2.228)	2	6.67
Chlordane cis	0.425 ± 0.162 (<LOD–0.893)	6	20.00		0	0.00	0.050 ± 0.009 (<LOD–0.058)	2	6.67	0.237	1	3.33
Chlordane trans		0	0.00		0	0.00		0	0.00	0.043 ± 0.010 (<LOD–0.059)	3	10.00
Endosulfan I		0	0.00	2.113 ± 1.095 (<LOD–3.208)	2	7.14		0	0.00		0	0.00
Aldrin	2.988	1	3.33		0	0.00	1.839	1	3.33	0.255 ± 0.037 (<LOD–0.489)	12	40.00
Endrin	0.631 ± 0.281 (<LOD–3.701)	13	43.33	1.710 ± 0.254 (<LOD–4.325)	18	64.29	0.253 ± 0.046 (<LOD–0.372)	5	16.67	0.188 ± 0.046 (<LOD–0.299)	4	13.33
Dieldrin		0	0.00		0	0.00		0	0.00	0.854 ± 0.595 (<LOD–2.041)	3	10.00
PCB-118		0	0.00	0.604 ± 0.124 (<LOD–0.727)	2	7.14		0	0.00		0	0.00
PCB-101		0	0.00	0.705 ± 0.153 (<LOD–1.427)	9	32.14		0	0.00		0	0.00
PCB-138		0	0.00	0.560 ± 0.216 (<LOD–1.591)	6	21.43		0	0.00		0	0.00

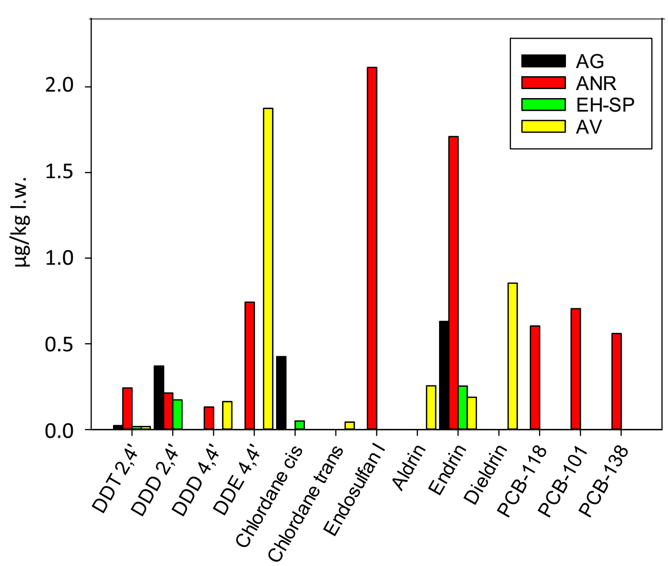

Fig. 2. Chlorine organic pollutants in muscle samples of European eels, *Anguilla anguilla*, in the four studied ecosystems. Places with only one specimen (AG and EH-SP for Aldrin, AV for DDD-2,4′ and chlordane cis) were no considered.

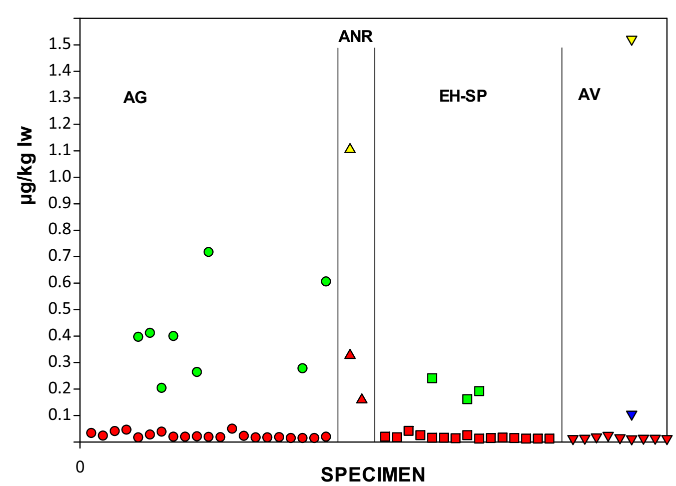

Fig. 3. Concentrations of metabolites in specimens of European eel *Anguilla anguilla* with DDT 2,4′ > LOD. Red: DDT 2,4′; green: DDD 2,4′; blue: DDD 4,4′; yellow: DDE 4,4′. AG = S'Albufera des Grau Natural Park; ANR = S'Albufereta Natural Reserve; EH-SP = El Hondo-Salinas de Santa Pola Natural Park; AV = Albufera de Valencia. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

both data (percentages vs. concentrations) should be taken into account. This relative abundance of DDT in eel muscles could therefore reflect their low metabolic capacity, ease of access, and/or a limited excretion capacity of DDT (Bonnineau et al., 2016). However, it must be taken into account that DDT was used as an intermediate product in the synthesis of dicofol (Rasenberg and van de Plassche, 2002), a banned (Commission Delegated Regulation (EU) 2020/1204) pesticide manufactured in Spain until 2008 and detected in water samples from a near Mediterranean ecosystem (Ebro River Delta) (Barbieri et al., 2021).

In the whole population, the usual correlation between DDTs and metabolites concentration of the fishes and their weight or length (Larsson et al., 1991; Madenjian et al., 1994) was not found. In this sense, other authors reporting no relationships (de la Cal et al., 2008; Nzau Matondo et al., 2022), biomagnification (Deribe et al., 2013) and negative correlation (Feng et al., 2022). According the last authors, this could be due to the bio-dilution of these chemicals during the fish growth process, and they highlight that the larger/older fish may eliminate these pollutants for a longer period of time compared to smaller/younger ones. However, this relationship was no observed in eels from each ecosystem, so this should be monitored on a regular basis, to permit a better understanding of the relation between these pollutants and the eels populations.

In three of the studied areas (AG, EH-SP and AV), the mean concentrations of DDT 2,4′ were similar and low (0.02 μg/kg, l.w.), while higher mean concentrations of DDD and DDE than DDT were recorded (Table 3). Again, it seems there was a close relationship between DDT and its metabolites, although only 18.2 (AV), 20 (EH-SP) and 38.1 % (AG) of specimens with DDT 2,4′ > LOD had DDD and/or DDE in muscles. In the South of the province of Alicante, Gómez-Ramírez et al. (2019) reported the decreasing concentrations of OCPs due to European OCPs regulation, and Deudero et al. (2007) show a general decrease in all species of human consumption. In our study, the highest mean concentrations of DDT (0.24 μg/kg, l.w.) were found in ANR (in only two specimens were found to contain this compound). In AV, DDT contamination has been reported in the past (Peris et al., 2005). However, Campillo et al. (2017) reported the existence of fresh inputs of DDT in the Valencia area, probably associated with high precipitation, leading to an increase in the transport of DDTs associated with sediments and particulate matter from rivers into the Mediterranean Sea. In addition, an area near this lake is cultivated for rice, which could explain these results. In 2008, the ecological status of AG and ANR were regarded as good (Plan Hidrológico de las Illes Balears 2015–2021), although some threats were reported to exist (e.g. wastewater from a water-treatment plant, infiltration from septic tanks and slurry from pig farms). However, it seems that eels from AG are more threatened than those from ANR in terms of DDT 2,4′ and DDD 2,4′ (70.0 and 33.3 % of specimens, respectively from AG; 7.1 % from ANR), since the high mean concentration of DDT 2,4′ in ANG was only found in two specimens (Table 3). Curiously, the main economic sector in these areas is tourism, and agriculture is only a secondary activity and there has been a steady increase in the amount of organic farming (CBPAE, 2020). In addition, to the best of our knowledge, no data regarding OCPs in these areas has been found other than in children, where high levels of DDE 4,4′ were reported, possibly due to the intense use of DDT in the past (Gascon et al., 2015).

Dieldrin is commonly found in biotic and abiotic matrices partly due to its stability (Jorgenson, 2001). However, this compound, which accumulates in wild species, and whose ubiquity and long-term existence as an environmental pollutant have been reported (see review in Pang et al., 2022), was only detected in 2.5 % of the samples (three specimens from AV). In our study, the mean detected values were similar to those quantified in southern France (Roche et al., 2003) but markedly lower than values determined in eel muscle samples from Belgium (5.70–15.6 μg/kg) (Maes et al., 2008; Van Ael et al., 2014), Italy (8.48–12.02 μg/kg) (Ferrante et al., 2010) and Ireland (1.4–3.5 μg/kg) (McHugh et al., 2010), which thus confirms the decrease of dieldrin in aquatic systems in recent decades (Tabouret et al., 2011). The use of dieldrin has been prohibited since 1974, while the use of its metabolite endrin has never been authorised (Maes et al., 2008). Endrin can be rapidly metabolized by animals (reviewed by Zitko et al., 2003) and does not accumulate to any great extent in lipids. Thus, it is not detected at high concentrations, except in recent exposures (Smith, 1991). In comparison to dieldrin, we detected endrin in 33 % of the samples, with a mean value closely related to that quantified in Belgian eel samples (Maes et al., 2008). Like dieldrin, endrin levels seem to have decreased in recent decades (see review in Ingber et al., 2021), which demonstrates that bans and environmental policies do lead to decreased concentrations. This compound was present in a high percentage of samples from the Balearic Islands (64.3 and 43.3 % in ANR and AG, respectively), with significantly high concentrations also in ANR; thus, the origin of this compound (and others including DDTs and their metabolites) in specimens caught in these areas should be investigated. Finally, aldrin was found mainly in AV (Table 3), the only area in which all three compounds were present. Peris et al. (2005), however, did not detect dieldrin and endrin in most of the sediments from AV and only very low concentrations of aldrin, findings that agree with our results. This compound can be metabolized by epoxidation in the liver to dieldrin (reviewed by Zitko, 2003), so its presence in this ecosystem should also be investigated.

Chlordane and endosulfan were detected in low percentages (Table 2). In the former, the isomer cis- was the major constituent, followed by trans-; nevertheless, in all cases they were present in <10 % of the whole population. This result agrees with those of Tabouret et al. (2011), who did not detect either of these two contaminants in any of the eel samples from French (Tabouret et al., 2011) and Belgian (Hoff et al., 2005) wetlands.

4.2. Polychlorinated biphenyls

The mean levels of marker-PCBs in our study ranged from 0.56 to 0.70 μg/kg (Table 2). In general, PCBs 118, 101 and 138 were characterized by their low detection frequency, with percentages in all cases below 10 % in the analyzed specimens. Curiously, eels with these compounds were only found at one site (ANR). It is interesting to note that in Europe, these PCBs, together with PCB 153 (four of the seven 'targets' recommended by the European Union for assessing PCB contamination), are the most dominant congeners in eels, although the relative abundance of individual congeners in specimens varies in terms of their origin and country considered (Belpaire et al., 2011b). For example, Flemish eels are mostly associated with higher proportions of congeners 153 and 180 compared to other European regions. In a study carried out in two Italian coastal lagoons (Caprolace and Fogliano), the two congeners that mostly contributed to the total PCB load were 153 and 138 (Leone et al., 2020); in another study carried out in southern Italy (Ferrante et al., 2010), the four most dominant congeners were 138, 153, 118 and 180, and at higher proportions (22.9, 18.9, 12.4 and 10.0 %, respectively) than those obtained in our study. A similar result was obtained in Germany, with the same congeners being the most abundant in eel samples (Fromme et al., 1999). In a study of eels from the river Turia (eastern Spain), congener 138 was the most abundant, although congeners 180 and 118 were also highly relevant (Bordajandi et al., 2003). It is interesting to note that long-term monitoring studies of eels in the Netherlands (since 1977) and Belgium (since 1994) indicate that overall there has been a significant decline in PCB levels (Malarvannan et al., 2014). Moreover, these studies indicate that in Europe, PCBs 153 and 138 are the most dominant congeners in eels, although the relative abundance of individual congeners in samples depends on the origin and country considered.

Despite the prohibitions that have existed since the 1970s on the use of these compounds, it takes decades for their quantities to decrease in the environment; they are stable in the environment for a long time, and they become widespread via water and atmospheric transport mechanisms (Korytár et al., 2002; Villa et al., 2003).

On the other hand, no relationships were found between PCBs concentration and biometric data. According to Couderc et al. (2015), the PCBs levels are usually correlated to lipid weight. Nevertheless, some authors have reported a positive correlation between total length and PCB-180 and none in remaining PCBs (Nzau Matondo et al., 2022). The relationship between these organic compounds and biological data such as fish length have been subjects of many studies (Green and Knutzen, 2003; Pikkarainen and Parmanne, 2006; Pandelova et al., 2008; Polak-Juszczak et al., 2022), with increase in older specimens by changes in feeding habits and longer exposure times (Kiviranta et al., 2003; Pandelova et al., 2008). The influence of fat content in the levels of these pollutants with a lack of correlation between the size of the fishes and the lipid content could be an explanation of this absence of correlations (de la Cal et al., 2008).

Surprisingly, these compounds were only detected in one of the two areas from the Balearic Islands. Mallorca has approximately 925,000 inhabitants, while Menorca has <100,000; there is approximately 3640 ha of agricultural land on Mallorca, whereas Menorca there is only about 701 ha (Govern de les Illes Balears, 2016). As well, there are also discharges of residual water from the refrigerating systems of thermal power plants, which use water with several chemical compounds including organochlorines. Although these compounds are used in low concentrations, the accumulated pollution due to the amount of water discharged could be relevant (EPA, 2009). In addition, the amount of refrigerated water discharged into the ocean from Menorca is 2.91 % of the water discharged from Mallorca (Govern de les Illes Balears, 2016). Thus, all these factors probably play a role in the different contamination levels of PCBs in eels from ANR and AG. Curiously, in samples of birds collected during the period 1994–2000, Jiménez et al. (2007) reported high PCBs levels in eggs of osprey (*Pandion haliaetus*) from Menorca, a species in the high trophic-level predators, that feed almost exclusively on fish such as saddled seabream (*Oblada melanura*) and mullet (Mugil sp.) (Triay, 1995).

On the other hand, the absence of positive specimens of PCBs from AV could be unusual, since the proximity of both industrial and urban centres to this site could affect the presence of these compounds in the eels from the area, which was subjected to great environmental pressure in the 1960s and 1970s (Segarra Ferrando and Dies Jambrino, 2014). A study performed in the Region of Valencia in 2010–2011 (Quijano et al., 2018) showed that the exposure of the general population to PCBs have declined by the efficiency of the European risk-management measures, so the null presence in eels would seem to be a good indicator for this family of compounds.

5. Conclusion

COPs are found in all hydrosystems due to the impact of every-day human activities (e.g. agricultural pollutants and industrial activities) and the persistence of the compounds used in the past (Bordajandi et al., 2003; Arai, 2014). At the population level, the possibility of a real threat due to COPs in eels from study areas would be low: 55.9 % of specimens had one or none compounds, only 18 eels (15.3 %) had more of 2 COPs, and only one eel presents 7 COPs (3 OCPs and 4 PCBs). Nevertheless, we found differences between the sampling sites in the anthropological impact of contaminants, as well as dissimilar management actions aimed at protecting the environment. Although these differences could explain some of the results of our study, other questions such as the high percentage of DDT 2,4′ in AG, an ecosystem recognized as having a good ecological status, or the presence of PCBs in ANR, require further study.

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CRediT authorship contribution statement

Alonso Pérez-Vegas: Conceptualization, Data curation, Formal analysis, Investigation, Writing – original draft. **Marcos Pérez-López:** Formal analysis, Investigation, Writing – review & editing. **Elena Barcala:** Data curation, Formal analysis, Investigation, Writing – review & editing. **Diego Romero:** Conceptualization, Data curation, Formal analysis, Resources, Supervision, Validation, Writing – review & editing. **Pilar Muñoz:** Conceptualization, Funding acquisition, Project administration, Resources, Supervision, Validation, Writing – review & editing.

Declaration of competing interest

None.

Data availability

The data that has been used is confidential.

References

Arai, T., 2014. Variation in organochlorine accumulation in relation to the life history of the Japanese eel Anguilla japonica. Mar. Pollut. Bull. 80 (1–2), 186–193. https://doi.org/10.1016/j.marpolbul.2014.01.011.

ATSDR, 2020. Toxicological profile for polychlorinated biphenyls (PCBs). Agency for Toxic Substances and Disease Registry (24 March 2022). https://wwwn.cdc.gov/TSP/ToxProfiles/ToxProfiles.aspx?id=142&tid=26.

Barbieri, M.V., Peris, A., Postigo, C., Moya-Garcés, A., Monllor-Alcaraz, L.S., Rambla-Alegre, M., Eljarrat, E., López de Alda, M., 2021. Evaluation of the occurrence and fate of pesticides in a typical Mediterranean delta ecosystem (Ebro River Delta) and risk assessment for aquatic organisms. Environ. Pollut. 274, 115813 https://doi.org/10.1016/j.envpol.2020.115813.

Beliaeff, B., Oconnor, T.P., Daskalakis, D.K., Smith, P.J., 1997. US mussel watch data from 1986 to 1994: temporal trend detection at large spatial scales. Environ. Sci. Technol. 31, 1411–1415.

Bellehumeur, K., Lapointe, D., Cooke, S.J., Moon, T.W., 2016. Exposure to sublethal levels of PCB-126 impacts fuel metabolism and swimming performance in rainbow trout. Comp. Biochem. Physiol. B Biochem. Mol. Biol. 199, 97–104. https://doi.org/10.1016/j.cbpb.2016.01.005.

Belpaire, C., Goemans, G., 2007. The European eel Anguilla Anguilla, a rapporteur of the chemical status for the water framework directive? Vie Milieu 57 (4), 235–252.

Belpaire, C., Geeraerts, C., Evans, D., Ciccotti, E., Poole, R., 2011a. The european eel quality database: towards a pan-european monitoring of eel quality. Environ. Monit. Assess. 183 (1–4), 273–284.

Belpaire, C., Geeraerts, C., Roosens, L., Neels, H., Covaci, A., 2011. What can we learn from monitoring PCBs in the European eel? A Belgian experience. Environ. Int. 37 (2), 354–364. https://doi.org/10.1016/j.envint.2010.10.006.

Bonnineau, C., Scaion, D., Lemaire, B., Belpaire, C., Thomé, J.P., Thonon, M., Leermaker, M., Gao, Y., Debier, C., Silvestre, F., Kestemont, P., Rees, J.F., 2016. Accumulation of neurotoxic organochlorines and trace elements in brain of female european eel (Anguilla anguilla). Environ. Toxicol. Pharmacol. 45, 346–355. https://doi.org/10.1016/j.etap.2016.06.009.

Bordajandi, L.R., Gómez, G., Fernández, M.A., Abad, E., Rivera, J., González, M.J., 2003. Study on PCBs, PCDD/Fs, organochlorine pesticides, heavy metals and arsenic content in freshwater fish species from the river turia (Spain). Chemosphere 53 (2), 163–171. https://doi.org/10.1016/S0045-6535(03)00417-X.

Brown, S.B., Adams, B.A., Cyr, D.G., Eales, J.G., 2004. Contaminant effects on the teleost fish thyroid. Environ. Toxicol. Chem. 23 (7), 1680–1701. https://doi.org/10.1897/03-242.

Campillo, J.A., Fernandez, B., García, V., Benedicto, J., León, V.M., 2017. Levels and temporal trends of organochlorine contaminants in mussels from spanish Mediterranean waters. Chemosphere 182, 584–594.

Carro, N., Garcia, I., Ignacio, M., Mouteira, A., 2004. Possible influence of lipid content on levels of organochlorine compounds in mussels from Galicia coast (Northwestern, Spain). Spatial and temporal distribution patterns. Environ. Int. 30, 457–466.

CBPAE, 2020. Consell Producció Agrària Ecològica. Dades estadístiques de la producció agrària ecològica Illes Balears 2020. http://www.cbpae.org/files/EAE_2020.pdf. (Accessed 1 April 2022).

CHJ-GV-AV, 2019. Borrador del Plan especial de l'Albufera. Confederación Hidrográfica del Júcar, Generalitat Valenciana y Ajuntament de València. Febrero de 2019.

CHS, 2020. Confederación Hidrográfica del Segura, O.A. Comisaría de Aguas. Desarrollo del Programa de Seguimiento para determinar el Estado de las Aguas Continentales y el Control de las Zonas Protegidas en la Demarcación Hidrográfica del Segura. Informe Final. Campaña 2019. Ministerio para la Transición Ecológica y el Reto Demográfico.

Carles, L., Martin-Laurent, F., Devers, M., Spor, A., Rouard, N., 2021. Potential of preventive bioremediation to reduce environmental contamination by pesticides in an agricultural context: a case study with the herbicide 2,4-D. J. Hazard. Mater. 416, 125740.

Commission Delegated Regulation (EU), 2020. 1204 of 9 June 2020 amending Annex I to Regulation (EU) 2019/1021 of the European Parliament and of the Council as regards the listing of dicofol. Off. J. Eur. Union L270, 4–6.

Couderc, M., Poirier, L., Zalouk-Vergnoux, A., Kamari, A., Blanchet-Letrouvé, I., Marchand, P., Vénisseau, A., Veyrand, B., Mouneyrac, C., Le Bizec, B., 2015. Occurrence of POPs and other persistent organic contaminants in the european eel (Anguilla anguilla) from the loire estuary, France. Sci. Total Environ. 505, 199–215. https://doi.org/10.1016/j.scitotenv.2014.09.053.

de la Cal, A., Eljarrat, E., Raldúa, D., Durán, C., Barceló, D., 2008. Spatial variation of DDT and its metabolites in fish and sediment from Cinca River, a tributary of Ebro River (Spain). Chemosphere 70 (7), 1182–1189. https://doi.org/10.1016/j.chemosphere.2007.08.036.

Deribe, E., Rosseland, B.O., Borgstrøm, R., Salbu, B., Gebremariam, Z., Dadebo, E., Skipperud, L., Eklo, O.M., 2013. Biomagnification of DDT and its metabolites in four fish species of a tropical lake. Ecotoxicol. Environ. Saf. 95, 10–18. https://doi.org/10.1016/j.ecoenv.2013.03.020.

Deudero, S., Box, A., March, D., Valencia, J.M., Grau, A.M., Tintore, J., Calvo, M., Caixach, J., 2007. Organic compounds temporal trends at some invertebrate species from the Balearics, Western Mediterranean. Chemosphere 68 (9), 1650–1659. https://doi.org/10.1016/j.chemosphere.2007.03.070.

EPA, 2009. Steam Electric Power Generating Point Source Category: Final Detailed Study Report. United States Environmental Protection Agency, Washington, D.C.

European Commission, 2007. Council Regulation (EC) N° 1100/2007 of 18 September 2007 establishing measures for the recovery of the stock of European eel. Off. J. Eur. Union L248, 17–23.

European Communities, 1976. Directive 1976/464/ECC concerning pollution caused by dangerous substances discharged into the aquatic environment. Off. J. Eur. Communities L129.
European Communities, 2000. Directive 2000/60/EC of 23 October 2000 Establishing a Framework for Community action in the field of water policy. Off. J. L 327, 0001–0073.
Feng, W.L., Wu, J.P., Li, X., Nie, Y.T., Xu, Y.C., Tao, L., Zeng, Y.H., Luo, X.J., Mai, B.X., 2022. Bioaccumulation and maternal transfer of two understudied DDT metabolites in wild fish species. Sci. Total Environ. 20 (818), 151814 https://doi.org/10.1016/j.scitotenv.2021.151814.
Ferrante, M.C., Clausi, M.T., Meli, R., Fusco, G., Naccari, C., Lucisano, A., 2010. Polychlorinated biphenyls and organochlorine pesticides in european eel (Anguilla anguilla) from the Garigliano River (Campania region, Italy). Chemosphere 78 (6), 709–716. https://doi.org/10.1016/j.chemosphere.2009.11.026.
Fromme, H., Otto, T., Pilz, K., Neugebauer, F., 1999. Levels of synthetic musks; bromocyclene and PCBs in eel (Anguilla anguilla) and PCBs in sediment samples from some waters of Berlin/Germany. Chemosphere 39 (10), 1723–1735. https://doi.org/10.1016/S0045-6535(99)00066-1.
Gascon, M., Vrijheid, M., Garí, M., Fort, M., Grimalt, J.O., Martinez, D., Torrent, M., Guxens, M., Sunyer, J., 2015. Temporal trends in concentrations and total serum burdens of organochlorine compounds from birth until adolescence and the role of breastfeeding. Environ. Int. 74, 144–151.
Geeraerts, C., Belpaire, C., 2010. The effects of contaminants in european eel: a review. Ecotoxicology 19 (2), 239–266.
Gómez-Ramírez, P., Pérez-García, J.M., León-Ortega, M., Martínez, J.E., Calvo, J.F., Sánchez-Zapata, J.A., Botella, F., María-Mojica, P., Martínez-López, E., García-Fernández, A.J., 2019. Spatiotemporal variations of organochlorine pesticides in an apex predator: influence of government regulations and farming practices. Environ. Res. 176, 108543 https://doi.org/10.1016/j.envres.2019.108543.
Govern de les Illes Balears, 2015. Plan Hidrológico de las Illes Baleares (2015–2021). Memoria, Conselleria d'Agricultura, Medi Ambient i Territori. Palma. http://www.caib.es/sites/aigua/ca/pla_hidrolagic_de_les_illes_balears/.
Govern de les Illes Balears, 2016. Informe del Estado del Medio Ambiente en Baleares (Informe de Coyuntura 2014-2015). Consellería Medi Ambient, Agricultura i Pesca. Direcció General Educació Ambiental, Qualitat Ambiental i Residus. Palma. https://www.caib.es/sites/informesmediambient/es/informe_2014_-_2015_coyuntura/.
Green, N.W., Knutzen, J., 2003. Organohalogens and metals in marine fish and mussels and some relationships to biological variables at reference localities in Norway. Mar. Pollut. Bull. 46, 362–377. https://doi.org/10.1016/S0025-326X(02)00515-5.
Henry, T.B., 2015. Ecotoxicology of polychlorinated biphenyls in fish—a critical review. Crit. Rev. Toxicol. 45 (8), 643–661. https://doi.org/10.3109/10408444.2015.1038498.
Hernández, F., Portolés, T., Pitarch, E., López, F.J., Beltrán, J., Vázquez, C., 2005. Potential of gas chromatography coupled to triple quadrupole mass spectrometry for quantification and confirmation of organohalogen xenoestrogen compounds in human breast tissues. Anal. Chem. 77 (23), 7662–7672. https://doi.org/10.1021/ac050874+.
Hoff, P.T., Van Campenhout, K., Van De Vijver, K., Covaci, A., Bervoets, L., Moens, L., Huyskens, G., Goemans, G., Belpaire, C., Blust, R., De Coen, W., 2005. Perfluorooctane sulfonic acid and organohalogen pollutants in liver of three freshwater fish species in Flanders (Belgium): relationships with biochemical and organismal effects. Environ. Pollut. 137 (2), 324–333. https://doi.org/10.1016/j.envpol.2005.01.008.
Hugla, J.L., Thomé, J.P., 1999. Effects of polychlorinated biphenyls on liver ultrastructure, hepatic monooxygenases, and reproductive success in the barbel. Ecotoxicol. Environ. Saf. 42 (3), 265–273. https://doi.org/10.1006/eesa.1998.1761.
ICES, 2018. Report of the Joint EIFAAC/ICES/GFCM Working Group on Eels (WGEEL), 5–12 September 2018, Gdansk, Poland. ICES CM 2018/ACOM:15, 050 pp.
ICES, 2021. Workshop on the future of eel advice (WKFEA). ICES Sci. Rep. 3 (13) https://doi.org/10.17895/ices.pub.5988, 67 pp.
Ingber, S.Z., Zaccaria, K., Ingerman, L., 2021. Toxicological profile for endrin. Agency for Toxic Substances and Disease Registry, United States. https://stacks.cdc.gov/view/cdc/104367.
Islam, F.U., Jalali, S., Shafqat, M.N., Shah, S.T.A., 2017. Endosulfan is toxic to the reproductive health of male freshwater fish, Cyprinion watsoni. Naturwissenschaften 104 (11–12), 104. https://doi.org/10.1007/s00114-017-1526-9.
Islam, M.S., Tanaka, M., 2004. Impacts of pollution on coastal and marine ecosystems including coastal and marine fisheries and approach for management: a review and synthesis. Mar. Pollut. Bull. 48, 624–649.
Jiménez, B., Merino, R., Abad, E., Rivera, J., Olie, K., 2007. Evaluation of organochlorine compounds (PCDDs, PCDFs, PCBs and DDTs) in two raptor species inhabiting a mediterranean island in Spain. Env. Sci. Pollut. Res. 14 (Special Issue 1), 61–68. https://doi.org/10.1065/espr2006.01.015.
Jorgenson, J.L., 2001. Aldrin and dieldrin: a review of research on their production, environmental deposition and fate, bioaccumulation, toxicology, and epidemiology in the United States. Environ. Health Perspect. 109 (1), 113–139. https://doi.org/10.1289/ehp.01109s1113.
Kelce, W.R., Lambright, C.R., Gray, L.E., Roberts, K.P., 1997. Vinclozolin and p, p'-DDE alter androgen-dependent gene expression: in vivo confirmation of an androgen receptor-mediated mechanism. Toxicol. Appl. Pharmacol. 142 (1), 192–200. https://doi.org/10.1006/taap.1996.7966.
Kiviranta, H., Vartiainen, T., Parmanne, R., Hallikainen, A., Koistinen, J., 2003. PCDD/Fs and PCBs in Baltic herring during the 1990s. Chemosphere 50 (9), 1201–1216. https://doi.org/10.1016/S0045-6535(02)00481-2.
Korytár, P., Leonards, P.E.G., de Boer, J., Brinkman, U.A.Th., 2002. High-resolution separation of polychlorinated biphenyls by comprehensive two-dimensional gas chromatography. J. Chromatogr. A 958 (1–2), 203–218.
Langer, P., 2010. The impacts of organochlorines and other persistent pollutants on thyroid and metabolic health. Front. Neuroendocrinol. 31 (4), 497–518. https://doi.org/10.1016/j.yfrne.2010.08.001.
Larsson, P., Hamrin, S., Okla, L., 1991. Factors determining the uptake of persistent pollutants in an eel population (Anguilla Anguilla L.). Environ. Pollut. 69, 39–50.
Leone, C., Capoccioni, F., Belpaire, C., Malarvannan, G., Poma, G., Covaci, A., Tancioni, L., Cont, M., Ciccotti, E., 2020. Evaluation of environmental quality of Mediterranean coastal lagoons using persistent organic pollutants and metals in thick-lipped grey mullet. Water 12 (12), 1–19.
Maes, J., Belpaire, C., Goemans, G., 2008. Spatial variations and temporal trends between 1994 and 2005 in polychlorinated biphenyls, organochlorine pesticides and heavy metals in European eel (Anguilla anguilla L.) in Flanders, Belgium. Environ. Pollut. 153 (1), 223–237. https://doi.org/10.1016/j.envpol.2007.07.021.
Malarvannan, G., Belpaire, C., Geeraerts, C., Eulaers, I., Neels, H., Covaci, A., 2014. Assessment of persistent brominated and chlorinated organic contaminants in the european eel (Anguilla anguilla) in Flanders, Belgium: levels, profiles and health risk. Sci. Total Environ. 482–483 (1), 222–233. https://doi.org/10.1016/j.scitotenv.2014.02.127.
Madenjian, C.P., Carpenter, S.R., Rand, P.S., 1994. Why are the PCB concentrations of salmonine individuals from the same lake so highly variable. Can. J. Fish. Aquat. Sci. 51, 800–807.
Martyniuk, C.J., Doperalski, N.J., Feswick, A., Prucha, M.S., Kroll, K.J., Barber, D.S., Denslow, N.D., 2016a. Transcriptional networks associated with the immune system are disrupted by organochlorine pesticides in largemouth bass (Micropterus salmoides) ovary. Aquat. Toxicol. 177, 405–416. https://doi.org/10.1016/j.aquatox.2016.06.009.
Martyniuk, C.J., Doperalski, N.J., Prucha, M.S., Zhang, J.L., Kroll, K.J., Conrow, R., Barber, D.S., Denslow, N.D., 2016b. High contaminant loads in Lake Apopka's riparian wetland disrupt gene networks involved in reproduction and immune function in largemouth bass. Comp. Biochem. Physiol. D: Genomics Proteomics 19, 140–150. https://doi.org/10.1016/j.cbd.2016.06.003.
Mateo, R., Millán, J., Rodríguez-Estival, J., Camarero, P.R., Palomares, F., Ortiz-Santaliestra, M.E., 2012. Levels of organochlorine pesticides and polychlorinated biphenyls in the critically endangered iberian lynx and other sympatric carnivores in Spain. Chemosphere 86 (7), 691–700. https://doi.org/10.1016/j.chemosphere.2011.10.037.
McHugh, B., Poole, R., Corcoran, J., Anninou, P., Boyle, B., Joyce, E., Barry Foley, M., McGovern, E., 2010. The occurrence of persistent chlorinated and brominated organic contaminants in the european eel (Anguilla anguilla) in irish waters. Chemosphere 79 (3), 305–313. https://doi.org/10.1016/j.chemosphere.2010.01.029.
Miller, M.J., Bonhommeau, S., Munk, P., Castonguay, M., Hanel, R., Mccleave, J.D., 2015. A century of research on the larval distributions of the Atlantic eels: a re-examination of the data. Biol. Rev. 90, 1035–1064.
Mills, L.J., Chichester, C., 2005. Review of evidence: are endocrine-disrupting chemicals in the aquatic environment impacting fish populations? Sci. Total Environ. 343, 1–34.
Neves, P.A., Colabuono, F.I., Ferreira, P.A.L., Kawakami, S.K., Taniguchi, S., Figueira, R.C.L., Mahiques, M.M., Montone, R.C., Bícego, M.C., 2017. Depositional history of polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) and polycyclic aromatic hydrocarbons (PAHs) in an Amazon estuary during the last century. Sci. Total Environ. 615, 1262–1270. https://doi.org/10.1016/j.scitotenv.2017.09.303.
Nzau Matondo, B., Delrez, N., Bardonnet, A., Vanderplasschen, A., Joaquim-Justo, C., Rives, J., Benitez, J.P., Dierckx, A., Séleck, E., Rollin, X., Ovidio, M., 2022. A complete check-up of european eel after eight years of restocking in an upland river: trends in growth, lipid content, sex ratio and health status. Sci. Total Environ. 807, 151020 https://doi.org/10.1016/j.scitotenv.2021.151020.
Olisah, C., Adams, J.B., Rubidge, G., 2021. The state of persistent organic pollutants in south african estuaries: a review of environmental exposure and sources. Ecotoxicol. Environ. Saf. 219, 112316 https://doi.org/10.1016/j.ecoenv.2021.112316.
Palstra, A.P., van Ginneken, V.J.T., Murk, A.J., van den Thillart, G.E.E.J.M., 2006. Are dioxin-like contaminants responsible for the eel (Anguilla anguilla) drama? Naturwissenschaften 93, 145–148. https://doi.org/10.1007/s00114-005-0080-z.
Pandelova, M., Henkelmann, B., Roots, O., Simm, Ja, RV, M.L., Benfenati, E., Schramm, K.W., 2008. Levels of PCDD/F and dioxin-like PCB in Baltic fish of different age and gender. Chemosphere 71 (2), 369–378. https://doi.org/10.1016/j.chemosphere.2007.08.050.
Pang, S., Lin, Z., Li, J., Zhang, Y., Mishra, S., Bhatt, P., Chen, S., 2022. Microbial degradation of aldrin and dieldrin: mechanisms and biochemical pathways. Front. Microbiol. 13, 713375 https://doi.org/10.3389/fmicb.2022.713375.
Pereira, V.M., Bortolotto, J.W., Kist, L.W., de Azevedo, M.B., Fritsch, R.S., Oliveira, R.L., Pereira, T.C.B., Bonan, C.D., Vianna, M.R., Bogo, M.R., 2012. Endosulfan exposure inhibits brain AChE activity and impairs swimming performance in adult zebrafish (Danio rerio). NeuroToxicology 33 (3), 469–475. https://doi.org/10.1016/j.neuro.2012.03.005.
Peris, E., Requena, S., De la Guardia, M., Pastor, A., Carrasco, J.M., 2005. Organochlorinated pesticides in sediments from the Lake Albufera of Valencia (Spain). Chemosphere 60 (11), 1542–1549.
Perugini, M., Cavaliere, M., Giammarino, A., Mazzone, P., Olivieri, V., Amorena, M., 2004. Levels of polychlorinated biphenyls and organochlorine pesticides in some edible marine organisms from the Central Adriatic Sea. Chemosphere 57, 391–400.

Pike, C., Crook, V., Gollock, M., 2020. Anguilla anguilla. The IUCN Red List of Threatened Species 2020: e.T60344A152845178. https://doi.org/10.2305/IUCN.UK.2020-2.RLTS.T60344A152845178.en (24 March 2022).
Pikkarainen, A.-L., Parmanne, R., 2006. Polychlorinated biphenyls and organochlorine pesticides in Baltic herring 1985–2002. Mar. Pollut. Bull. 52, 1299–1309. https://doi.org/10.1016/j.marpolbul.2006.05.022.
Polak-Juszczak, L., Waszak, I., Szlinder-Richert, J., Wójcik, I., 2022. Levels, time trends, and distribution of dioxins and polychlorinated biphenyls in fishes from the Baltic Sea. Chemosphere 306, 135614. https://doi.org/10.1016/j.chemosphere.2022.135614.
Quijano, L., Marín, S., Millan, E., Yusà, V., Font, G., Pardo, O., 2018. Dietary exposure and risk assessment of polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans and dioxin-like polychlorinated biphenyls of the population in the Region of Valencia (Spain). Food Addit. Contam. Part A Chem. Anal. Control Expo. Risk Assess. 35 (4), 740–749. https://doi.org/10.1080/19440049.2017.1414960.
Rasenberg, M., van de Plassche, E.J., 2002. Information dossier on DDT used for the production of Dicofol. Ministerie van VROM. Final Report.
Roche, H., Buet, A., Ramade, F., 2003. Characterization and validation of ecotoxicological biomarkers in an eel population exposed to persistent organic pollutants in the Vaccares lake, French National Reserve of Camargue | Mise en évidence et validation de biomarqueurs écotoxicologiques dans dans la population d' anguilles d' un etang de la reserve naturelle nationale de Camargue, le Vaccares, exposee a des polluants organiques persistants. Rev. Ecol. (Terre Vie) 58 (1), 127–141.
Segarra Ferrando, J., Dies Jambrino, B., 2014. In: El parc natural de l'Albufera. Un paisaje cultural cargado de historia. Bienes, paisajes e itinerarios/Revista ph Instituto Andaluz del Patrimonio Histórico, 85, pp. 54–77.
Smith, A.G., 1991. Chlorinated hydrocarbon insecticides. In: Hayes, W.J., Jr Jr., Laws E. R. (Eds.), Handbook of Pesticide Toxicology, Classes of Pesticides, Vol. 2. Academic Press, Inc, New York, pp. 731–915, 1991.
Schmidt, J., 1923. Breeding places and migration of the eel. Nature 111, 51–54. https://doi.org/10.1038/111051a0.
Szlinder-Richert, J., Usydus, Z., Pelczarski, W., 2010. Organochlorine pollutants in European eel (Anguilla anguilla L.) from Poland. Chemosphere 80, 93–99.
Tabouret, H., Bareille, G., Mestrot, A., Caill-Milly, N., Budzinski, H., Peluhet, L., Prouzet, P., Donard, O.F.X., 2011. Heavy metals and organochlorinated compounds in the European eel (Anguilla anguilla) from the Adour estuary and associated wetlands (France). J. Environ. Monit. 13 (5), 1446–1456. https://doi.org/10.1039/c0em00684j.
Triay, R., 1995. Reproduccion del águila pescadora (Pandion haliaetus) en la isla de Menorca (Mediterrano Occidental). Ardeola 42, 21–28.
UNEP, 2018. Stockholm Convention on Persistent Organic Pollutants (POPs). In: Revised in 2018. Secretariat of the Stockholm Convention.
Van Ael, E., Belpaire, C., Breine, J., Geeraerts, C., Van Thuyne, G., Eulaers, I., Blust, R., Bervoets, L., 2014. Are persistent organic pollutants and metals in eel muscle predictive for the ecological water quality? Environ. Pollut. 186, 165–171. https://doi.org/10.1016/j.envpol.2013.12.006.
van Ginneken, V., Bruijs, M., Murk, T., Palstra, A., Van den Thillart, G., 2009. The effect of PCBs on the spawning migration of European Silver Eel (Anguilla anguilla L.). In: Van den Thillart, G., Dufour, S., Rankin, J.C. (Eds.), Spawning Migration of the European Eel, Fish & Fisheries Series, vol 30. Springer, Dordrecht. https://doi.org/10.1007/978-1-4020-9095-0_15.
van Ginneken, V., Palstra, A., Leonards, P., Nieveen, M., van den Berg, H., Flik, G., Spanings, T., Niemantsverdriet, P., van den Thillart, G., Murk, A., 2009b. PCBs and the energy cost of migration in the European eel (Anguilla anguilla L.). Aquat. Toxicol. 92, 213–220.
Villa, S., Finizio, A., Diaz Diaz, R., Vighi, M., 2003. Distribution of organochlorine pesticides in pine needles of an oceanic island: The case of tenerife (Canary islands, Spain). Water Air Soil Pollut. 146 (1–4), 335–349. https://doi.org/10.1023/A:1023906306701.
Windsor, F.M., Pereira, M.G., Tyler, C.R., Ormerod, S.J., 2019. River organisms as indicators of the distribution and sources of persistent organic pollutants in contrasting catchments. Environ. Pollut. 255, 113144 https://doi.org/10.1016/j.envpol.2019.113144.
Zitko, V., 2003. Chlorinated pesticides: Aldrin, DDT, Endrin, Dieldrin, Mirex. In: Fiedler, H. (Ed.), Persistent Organic Pollutants. The Handbook of Environmental Chemistry, vol 3O. Springer, Berlin, Heidelberg. https://doi.org/10.1007/10751132_4.