MINISTERIE VAN MIDDENSTAND EN LANDBOUW – BESTUUR ONDERZOEK EN ONTWIKKELING

CENTRUM VOOR LANDBOUWKUNDIG ONDERZOEK - GENT

DEPARTEMENT ZEEVISSERIJ

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Informatieve nota over PCBs in vis

Voorkomen in het mariene milieu

PCBs (polychloorbiphenyl, 209 in totaal, congeneren genaamd) zijn een verzameling van chemische componenten met dezelfde basisstructuur (biphenyl). Deze basisstructuur is in meer of mindere mate gesubstitueerd met chloor. Het aantal substituties, de plaats en de positie t.o.v. de ringstructuur bepalen hun afzonderlijke specifieke kenmerken. PCBs en dioxines zijn zeer gelijkaardige producten. Dioxines ontstaan uit PCBs onder andere door onvolledige verbranding. Dioxines zijn veel giftiger dan PCBs, maar sommige PCBs (de planaire) benaderen wel de toxiciteit van het meest toxische dioxine TCDD.

PCBs worden en werden voornamelijk gebruikt in transformatoroliën omdat ze hittebestendig en electrisch niet-geleidend zijn. Ze werden ook verwerkt in plastics en verf. Ze hadden en hebben dus een enorm economisch belang in tegenstelling tot dioxines die echte contaminanten zijn.

Door onomzichtige behandeling van die oliën of van producten gecontamineerd met deze oliën zijn heel wat PCBs in het milieu terecht gekomen. Er zou volgens een Nederlandse schatting tussen 1 en 10% van de totate wereldproductie in het mariene milieu terechtgekomen zijn. Volgens dezelfde studie was in de beginjaren '90, naar schatting nog 60 tot 70% van de totale wereldproductie in gebruik in gesloten systemen. Nochtans is de productie in Japan gestopt in '73, in de USA in '76. In Europa is ze doorgegaan tot '87.

De contaminatie van het milieu, ook van het mariene milieu is gekend. Vandaar dat internationale organisaties (bvb OSPARCOM) alle Europese landen met kustwateren verplicht hebben om de PCB concentratie in het mariene milieu op te volgen. Deze bezorgdheid wordt mede ingegeven door de grote persistentie van PCBs in het milieu. Dit wil zeggen dat ze slechts zeer traag afgebroken worden en daardoor voortdurend recycleren om uiteindelijk in het mariene milieu terecht te komen. Dit is dus een historische contaminatie van het mariene milieu in tegenstelling

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tot wat recent gebeurd is in de dierlijke veeteeltsector. Het zal nog decennia duren vooraleer PCBs in het mariene milieu in belangrijke mate zijn geëlimineerd op voorwaarde dat er geen inputs meer zijn.

Voor de Belgische kust is de monitoring van het mariene milieu toevertrouwd aan het CLO-Departement Zeevisserij. Het mariene milieu wordt regelmatig bemonsterd en PCBs worden bepaald in sedimenten, kabeljauw lever, kabeljauw vlees, botvlees, mosselen en garnalen.

De bekomen data worden gerapporteerd aan ICES (International Council for the Exploration of the Sea). De organisatie houdt hiervan een databank bij o.a. in opdracht van OSPARCOM, die gedeeltelijk toegankelijk is over het internet. De gegevens worden gebruikt door OSPARCOM voor de 'Quality Status Report' en de 'Interministriële Conferenties' over de Noordzee.

Bij de verwerking van de data over een periode van 13 jaar (83-96) kwam tot uiting dat de PCBs concentratie in mariene organismen voor de Belgische kust een dalende trend vertonen (zie bijgevoegde figuur over PCBs in bot). De gegevens vanuit andere landen duiden eveneens op een dalende trend in het mariene milieu.

NORMEN

Het nut van normeren op vetbasis.

PCB concentraties worden meestal, wanneer de data gebruikt worden voor monitoring doeleinden, uitgedrukt op vetbasis. De reden hiertoe is dat PCBs in hoofdzaak vetoplosbare producten zijn en zich dus in de vetfase ophouden. Bovendien sluit men door te normeren op vetbasis een deel natuurlijke variatie uit waardoor regionale verschillen en trends nauwkeuriger kunnen bepaald worden. Het laat ook toe om organismen met sterk uiteenlopende vetgehalten met mekaar te vergelijken. Naar menselijke consumptie toe is het beter PCB concentraties uit te drukken op vers of droog gewicht. Voor visvlees is dit uitermate belangrijk omdat visvlees, althans voor heel wat vissoorten, vetarm is. Platvissen en kabeljauwachtigen bevatten bijvoorbeeld zelden meer dan 1% vet.

Normen voor vis

Er bestaan in België geen normen voor PCBs in vis. Er bestaan wel normen voor PCBs in het buitenland. (zie Tabel). Bij vergelijking van de data in de figuur en de tabel blijkt dat deze consumptienormen niet overschreden worden in mariene organismen. Bij de interpretatie van de

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normen moet wel rekening gehouden worden met het feit dat niet in alle landen hetzelfde aantal congeneren in de norm verwerkt zitten.

De dioxinecrisis in België

Tijdens de dioxine crisis in België werd er voor de som van 7 congeneren (dwz 7 verschillende PCBs) een limiet gesteld van 200µg/kg vet. Hierbij moet heel duidelijk gesteld worden dat deze PCB norm een omgerekende dioxinenorm is. Deze norm geldt ook enkel voor de specifieke contaminatie die is opgetreden in de periode januari – maart 1999 in de vleesproductieketen. Hoogstwaarschijnlijk zullen op EEG niveau, mede gestimuleerd door de Belgische crisis, nieuwe Europese normen worden uitgewerkt. Deze normen zullen niet noodzakelijk dezelfde zijn als diegene die nu in de dioxine crisis gehanteerd worden.
PCBs worden soms ook uitgedrukt in dioxinen-equivalenten. Daarbij worden bepaalde omrekeningsfactoren gebruikt die onder andere opgesteld werden door de WHO. In een recente Britse studie (http.//www.maff.gov.uk/food/ infsheet/1999) werd aangetoond hoe dioxine en PCBs zich verdelen over het dagelijks dieet. Slecht 5% van de dagelijkse opname van dioxine-equivalenten vindt zijn oorsprong in vis. Bij een grote viseter loopt dit op tot 15%. In een aansluitend advies van een geneeskundige commissie werd aanbevolen om wekelijks een portie (vette) vis te eten, aangezien verwacht wordt dat de voordelen hiervan opwegen (vis bevat veel polyonverzadigde vetzuren van een bepaald type, die o.a. bijdragen tot een sterk verminderd risico op hart en bloedvatenziekten) tegen de nadelen (contaminatie met PCBs).

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PCBS IN COD (*Gadus morhua*), FLOUNDER (*Platichthys flesus*), BLUE MUSSEL (*Mytilus edulis*) AND BROWN SHRIMP (*Crangon crangon*) FROM THE BELGIAN CONTINENTAL SHELF: RELATION TO BIOLOGICAL PARAMETERS AND TREND ANALYSIS.

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Abstract

PCB levels in cod, flounder, mussel and shrimp, covering a ten-year period, were assessed for temporal trends and their relation to biological parameters. A significant relation was found between the PCB levels on a wet weight basis and the total lipid content. Normalising on the total lipid content reduced the differences in PCB levels between the organisms and between different tissues within the organisms. A general downward trend was observed for the PCB levels on the Belgian continental shelf.

Introduction

Polychlorinated biphenyls (PCBs) are ubiquitous environmental contaminants, which have caused world-wide concern since the discovery of their presence in the environment by Jensen [1]. Their widespread occurrence in the marine environment and their toxic potential resulted in a number of international monitoring programmes such as the Co-ordinated Monitoring Programme of the International Council for the Exploration of the Seas (ICES) and the Joint Monitoring Programme (JMP) of the Oslo and Paris Commissions (OSPARCOM) [2]. These programmes aimed to assess the levels of PCB contamination in the marine environment with emphasis on human consumption and the overall quality of the marine ecosystem, and to investigate possible trends in PCB levels.

The Belgian Fisheries Research Station is measuring PCBs in marine samples since 1978. The data presented here cover an eleven years' period (1983-1993) and form a solid basis to investigate time trends. Trends in the PCB concentrations in cod, flounder, mussel and shrimp on the Belgian continental shelf in relation to biological parameters such as fat content, age, weight, length and sex are assessed.

Materials and Methods

Materials

All materials used for this work were of research grade quality. Standard solutions were prepared on a weight basis from pure compounds (> 99 % pure) or certified reference standards.

Sampling

Cod (*Gadus morhua*), flounder (*Platichthys flesus*) and shrimp (*Crangon crangon*) were collected by the institute, using beam trawling, on the Belgian continental shelf from 1983 to 1993. Twenty-five individuals per fish species were sampled 2-3 months prior to spawning and divided in five length classes between 214 and 905 mm for cod and 200 and 450 mm for flounder. Muscle tissue was analysed individually but livers were pooled per length class. Shrimp sample sizes comprised 100 individuals. Cooked tail muscle was isolated and divided in five subsamples. Mussels (*Mytilus edulis*) were harvested on three jetties along the Belgian coast and sorted per length class of 20-30 mm, 30-40 mm, 40-50 mm and > 50 mm. Total sample sizes were between 150 and 617 individuals. The mussels were left in settled seawater at room temperature for 24 hours. Subsequently the soft body was isolated for analysis. All samples were stored at -28 °C prior to analysis.

Chemical analysis

Extraction was based on total lipid extraction according to the method of Bligh and Dyer [3]. The extracted lipids firstly used for the determination of the fat content were redissolved in hexane, and the resulting solution was subsequently cleaned on a Florisil column [4,5]. Analyses were performed on a Carlo Erba 4160 gas chromatograph equipped with an electron capture detector and a 25 m SE-54 column (before 1990) or (from 1990 onwards) a 60 m DB-17 and a 60 m DB-5 column (internal diameter 0.25 mm, film thickness 0.25 µm). Prior to 1989, the PCB concentrations were calculated on the basis of comparison with eight PCB peaks of Aroclor 1260 [4]. These eight peaks corresponded with IUPAC nos. 101, 136, 147, 153, 138, 128, 180 and 170 [6]. Concentrations of individual congeners, viz. IUPAC nos. 28, 31, 52, 101, 105, 118, 138, 153, 156 and 180 [6], are determined since 1989 [5]. Prior to 1990, quality assurance consisted of the analysis of procedural blanks, reproducibility and repeatability tests, injection of standard solutions as unknowns, and analysis of samples with known concentrations. Since 1990, the analysis of a certified reference material (BCR CRM 349) has been added as a standard procedure.

Conversion of data calculated with the Aroclor standard.

Due to the lack of individual PCB congener concentrations, before 1989, all statistical analyses had to be performed on the total PCB concentrations. Since 1989 individual congener concentrations are calculated and summed to express the total PCB concentration (ΣPCB). However, the resulting sum is not equal to the concentration calculated on the basis of Aroclor 1260 (Aroclor concentration). A conversion or recalculation method was therefore developed. The conversion is based on the fact that the ratio between the total PCB concentrations calculated with both methods should remain constant if the PCB patterns are identical and the ratios of the individual peaks to the total peak pattern are constant. A conversion factor (CF) can then be calculated, which is given in Equation 1.

$$(1) \qquad CF = \frac{[Aroclor]}{\Sigma PCB}$$

with [Aroclor] = concentration based on Aroclor 1260 and ΣPCB = summed concentration of individual PCBs

Consequently the older Aroclor concentration can now be recalculated to give ΣPCB values. All ΣPCB referred to in this paper are either the sums of individual congeners or the concentrations recalculated as described above.

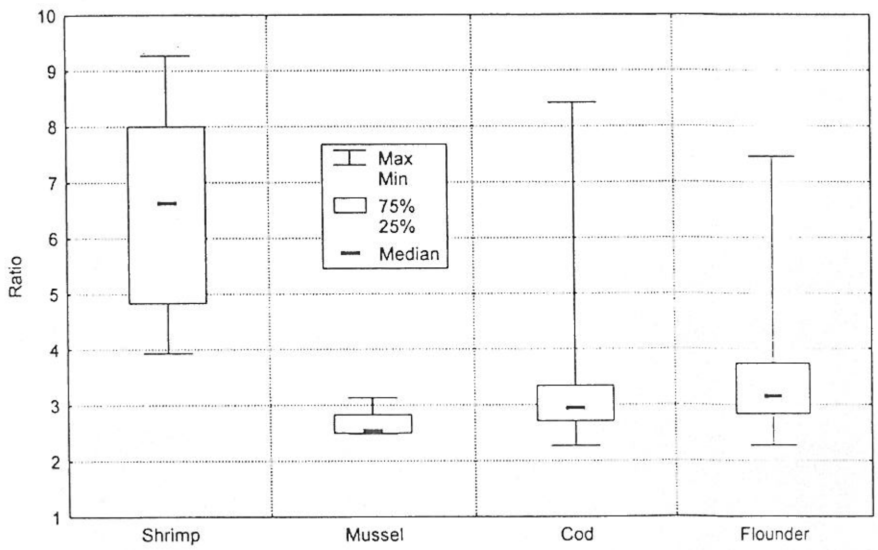

Figure 1: Ratio between ΣPCB and CB 153 for shrimp, mussel, cod and flounder in the period 1990-1993.

Statistical analysis

All statistical analyses were performed on ΣPCB and the level of significance was set at 95%. Correlations between fat content, length of the animal and PCBs were analysed by linear regression. The non-parametric Mann-Withney test was used to investigate the relation between sex and PCB concentrations, and the non-parametric Kruskall-Wallis ANOVA test combined with Dunns' post test was performed to compare the PCB contents of liver and muscle tissues in and among species and to study the influence of the age of the animals on the PCB content. Time-trend analysis of the PCB concentrations (median values per year) in cod, flounder and mussel were studied according to the method of Nicholson *et al.* [7]. PCB trends in shrimp were analysed by linear regression.

Results

Recalculation of the Aroclor concentrations

Conversing the Aroclor concentrations to ΣPCB by using Equation 1 requires that the PCB pattern of a given species/tissue is identical and that the ratios of the concentration of individual peaks to the total peak pattern are constant. The ratios between ΣPCB and CB 153 were calculated for the data obtained since 1990 and are presented by box and whisker plots in Figure 1. The results show a narrow box for cod, flounder and blue mussel and prove that the ratio remained constant; they also allow to suggest that the PCB patterns are similar. However, the shrimp data show a different pattern, which may find its origin in the rather small data set available. The PCB concentrations in the samples of cod and flounder, taken in 1991, and of mussel and shrimp, taken in 1991-1992, were then calculated using both methods of calculation and for each sample the CF was calculated according to Equation 1 (Table 1). Next, the Aroclor concentrations were re-calculated into ΣPCB.

Table 1: Conversion factors (CF) for the recalculation of 'Aroclor' data.

Species	CF
Cod	4.20 ± 0.04 (n=25)
Flounder	3.6 ± 0.1 (n=25)
Mussel	3.4 ± 0.1 (n=12)
Shrimp	3.1 ± 0.1 (n=10)

In principle, the CF value of 3.1 cannot be used to recalculate the Aroclor concentrations of shrimp, since the experimental results do not provide the required proof. However, the standard deviation of the CF is rather small; it is, moreover, similar to that of the other species. Moreover, a similarity between PCB patterns in invertebrates has been reported in the literature [8,9] and is indeed found for mussel (cf. above). We therefore assumed that the PCB patterns in the same species of invertebrate from the same location will be essentially the same and used the CF-based procedure also to recalculate the older data for shrimp.

Relation between PCB concentrations and total lipid content.

The results of the correlation analysis between the total lipid content and log (ΣPCB) for the different species and tissues are given in Table 2. A significant correlation ($p<0.05$) was found for the log (ΣPCB) expressed on a wet weight basis and the total lipid content, despite the large variability of the data (Figure 2). In contrast, no significant correlation was found when the concentrations were normalised on the total lipid content (Table 2).

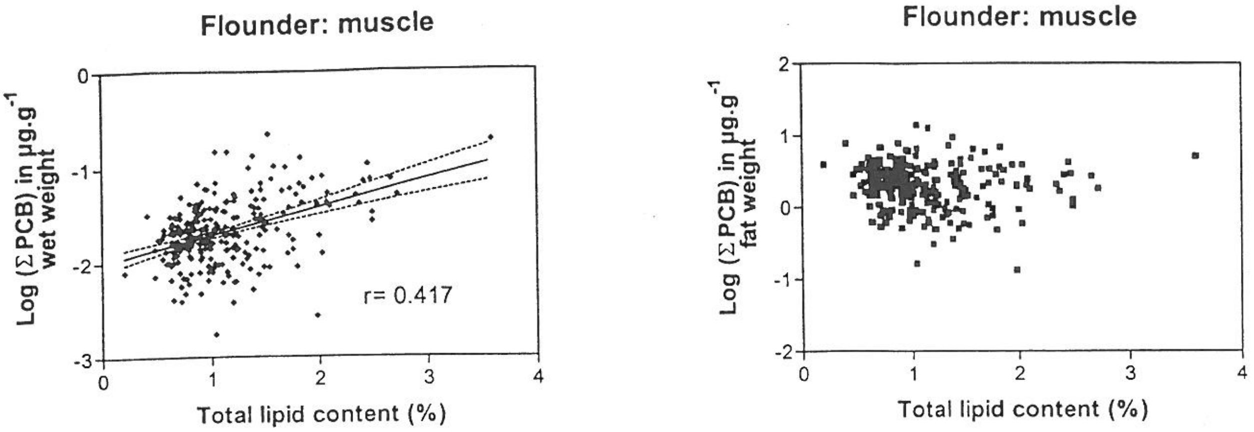

Figure 2: Relationship between total lipid content and log (ΣPCB) expressed on wet and fat weight basis (r, correlation coefficient; dotted line, 95% confidence interval of the mean).

The effect of lipid normalisation of the PCB data is illustrated in Figure 3 for all species and tissues examined. The results of a Kruskal-Wallis ANOVA analysis of the data indicate significant ($p<0.05$) inter-tissue and inter-species differences. However, narrowing this down with Dunn's post test revealed that the differences between cod liver, flounder liver, flounder muscle and blue mussel (soft body tissue) were not significant. Obviously, normalisation on the total lipid content reduces the differences in PCB levels between the organisms and between different tissues within the organisms, that is, the results illustrate the importance of lipids as a normalising factor. PCB concentrations are therefore only considered on a fat weight basis in the remainder of this paper.

Table 2: Results of correlation analysis between total lipid content and log (ΣPCB) on wet and fat weight basis for the different species and tissues.

Parameter	Total lipid content (%) vs. log (ΣPCB) on wet weight basis		Total lipid content (%) vs. log (ΣPCB) on fat weight basis	
	r	p	r	p
Cod muscle	0.25	<0.05	-	0.7255
Cod liver	-	-	-	-
Flounder muscle	0.42	<0.05	-	0.1392
Flounder liver	-	-	-	-
Blue mussel	0.50	<0.05	-	0.7526
Brown shrimp	0.51	<0.05	-	0.7685

p = p value, r = correlation coefficient

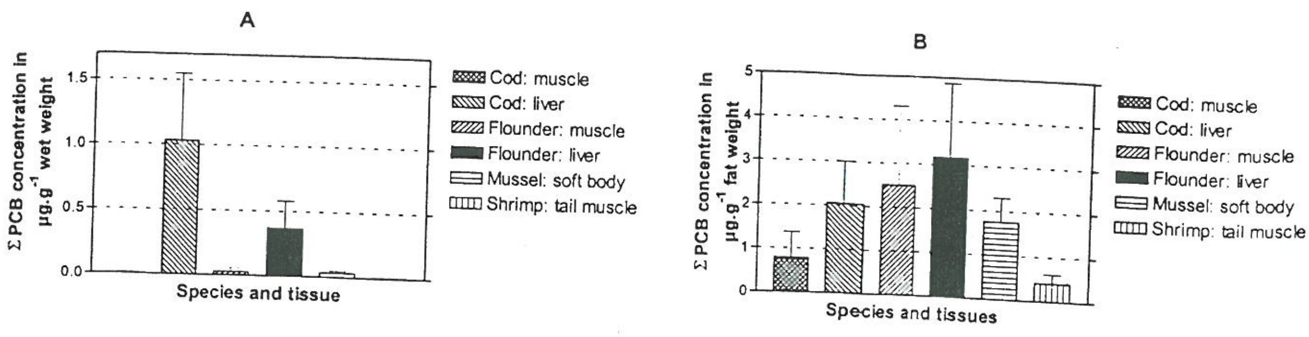

Figure 3: PCB concentrations for cod, flounder, mussel and shrimp, (A) not normalised and (B) normalised on total lipid content.

Relations between ΣPCB and length and sex

The trend analysis of Nicholson *et al.* [7] dictates a different approach when a length effect has been established. As regards both fish species, no demonstrable size effects were found except in cod liver (Figure 4). The ΣPCB concentrations were in addition to body size also related to sex, but no significant relations were found.

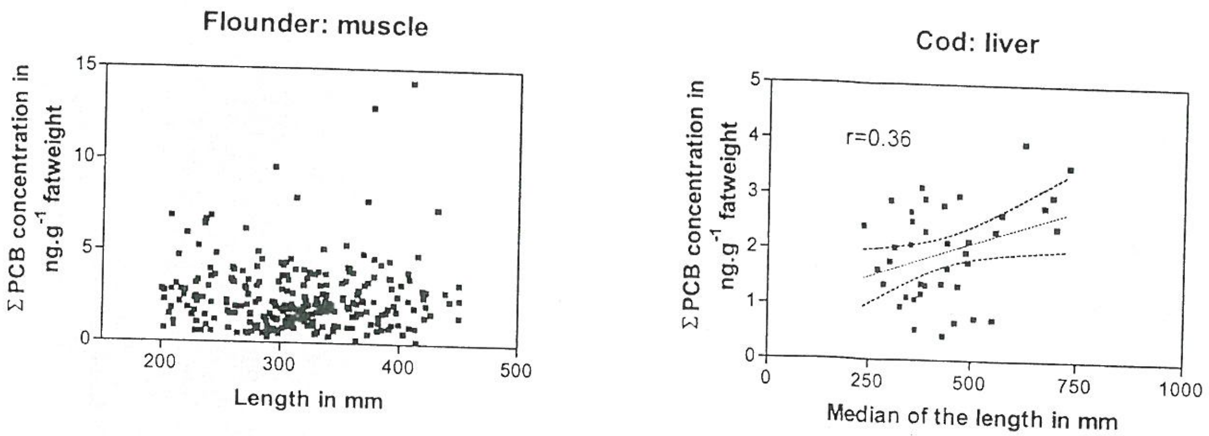

Figure 4: Relationship between length and log (ΣPCB) expressed on fat weight basis for flounder muscle tissue (left) and cod liver (right) (line illustrates calculated significant trends; r, correlation coefficient; dotted line, 95 % confidence interval of the mean).

As a result of the sampling procedure, no individual size data were available for the invertebrates. For mussel, however, samples were divided into five length classes and the ΣPCB concentrations were compared. The results are shown in Figure 5. The length class has, apparently, no effect on the ΣPCB concentrations, which was confirmed with a Kruskal-Wallis ANOVA test.

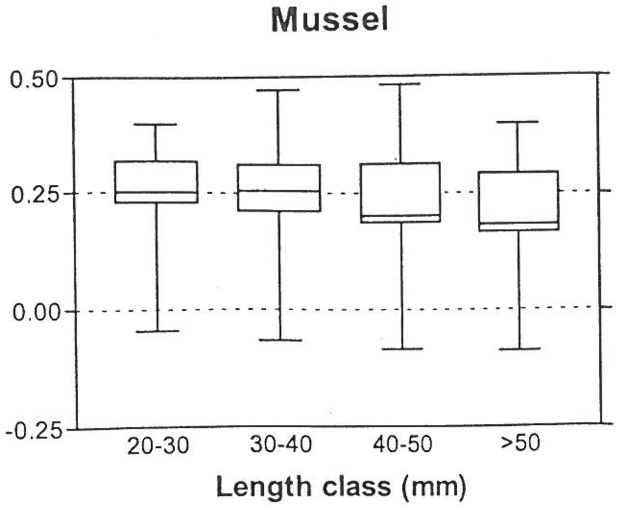

Figure 5: Relationship between length class and log (ΣPCB) expressed on a fat weight basis for blue mussel (box, median and 25 and 75 percentiles; whiskers, minimum and maximum).

Temporal-trend analysis

The observed absence of relations between the PCB content and the animals' length or sex allowed the analysis of temporal trends in cod muscle, flounder muscle and flounder liver tissues and in blue mussel without statistical modifications, but not for cod liver with which a length effect was found, nor for brown shrimp for which the length effect was not studied. As regards the cod liver data, they were subdivided at the median into a 'small' and a 'large' group and both were analysed independently [7]. Temporal trends in brown shrimp data were analysed by linear regression. The data were log transformed in order to approach the normal distribution. The temporal trends are illustrated in Figure 6 and the lipid normalised mean and median concentrations are given in Table 3. Long-term changes in the PCB concentrations were only considered as significant within a 95% confidence interval. The results revealed (1) significant year-to-year differences in cod and flounder muscle tissues and flounder liver tissue, (2) a significant downward non-linear trend in cod muscle, (3) a significant downward linear trend in flounder muscle, (4) no trend in blue mussel tissue, (5) a significant downward trend in brown shrimp and (6) no significant trends in cod and flounder liver tissues.

Discussion

For the four species studied the PCB concentrations expressed on a wet weight basis show a significant correlation with the fat content. This finding agrees well with previous observations. Schaefer *et al.* [10] demonstrated that PCB concentrations per wet weight in different tissues of cod rose with increasing lipid content as did Schneider [11]. Goerke *et al.* [12] found positive correlations between PCB concentrations and the fat content of various marine organisms. Positive correlations between PCB concentrations on a dry weight basis and the lipid content of various marine organisms were also reported by Delbeke *et al.* [13].

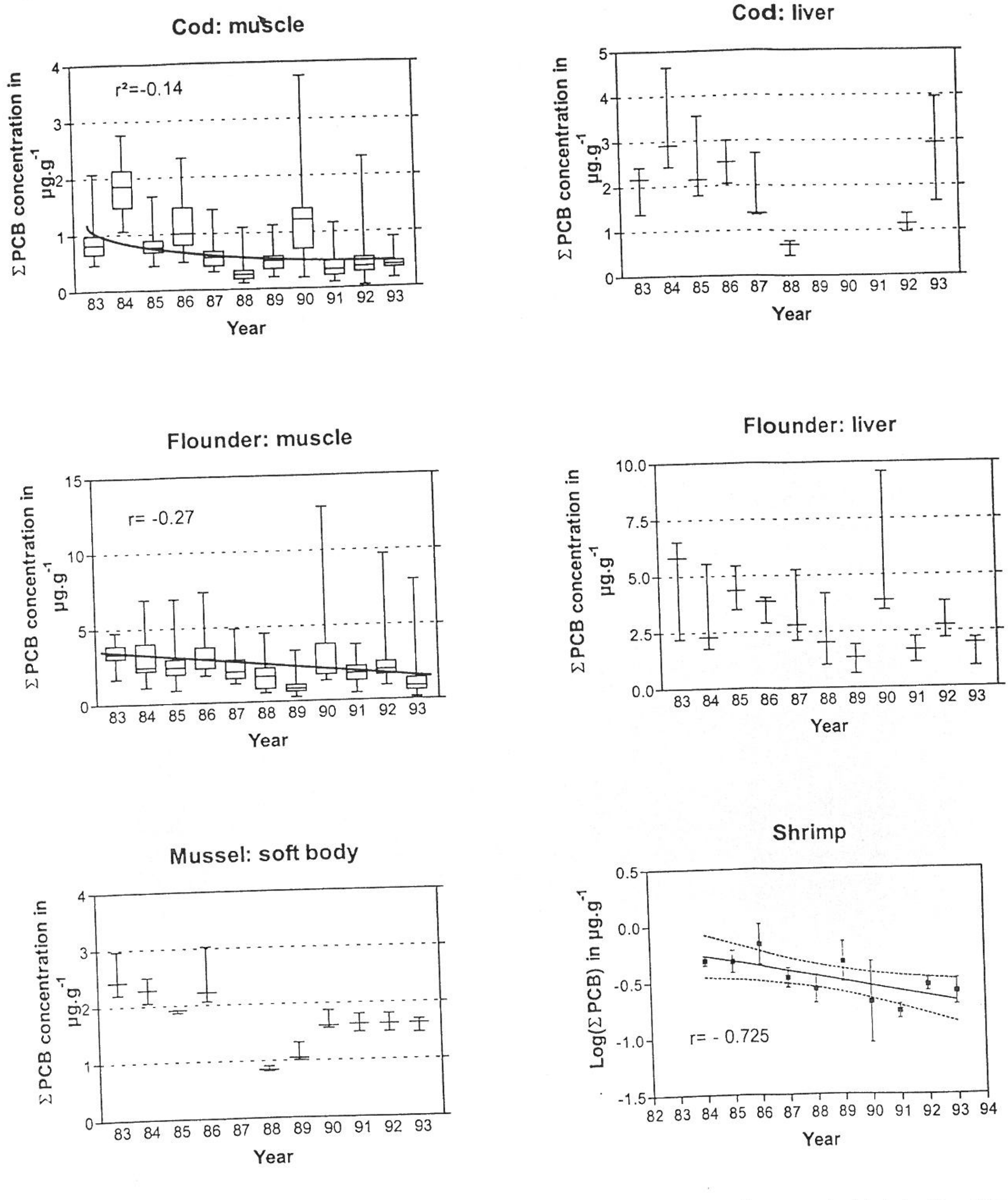

Figure 6: ΣPCB concentrations in µg/g fat weight for cod, flounder and mussel (lines illustrate calculated significant trends; r , correlation coefficient; boxes, median and 25 and 75 percentiles; whiskers, minimum and maximum) and linear trend for the log(ΣPCB) concentration in shrimp (r, correlation coefficient; dotted line, 95 % of the mean).

Moreover, inter-species and tissue-type differences decreased when PCB concentrations were normalised for the fat content. The correlation between fat content and log (ΣPCB) illustrates the need for a normalisation of the PCB concentrations on a fat basis, especially when a time-trend assessment is attempted. The explanation probably is that the natural variations in the lipid content of an organism or organ, due to e.g. spawning or lack of food, may influence the variability of contaminant data when these data are expressed on a fresh (wet) weight basis. Delbeke *et al.* [13] observed a similar reduction of the inter-species variability of PCB isomer concentrations after normalisation of the data on 'total neutral lipids', as determined by Iatroscan analysis. Using this selected class of lipids for normalisation proved superior to using the total lipid

Table 3: Mean and median concentration (µg/g fat weight), standard deviation (s) and number of samples (n) for the different species in the period 1983-1992.

	83	84	85	86	87	88	89	90	91	92	93
Cod muscle tissue											
Average	0.87	1.8	0.84	1.2	0.64	0.33	2.1	1.3	0.44	0.57	0.43
n	25	18	25	25	25	25	25	25	25	25	25
s	0.34	0.5	0.29	0.5	0.28	0.23	0.9	0.9	0.31	0.54	0.15
Median	0.81	1.9	0.77	1.0	0.58	0.26	2.1	1.2	0.33	0.38	0.40
Cod liver											
Average	2.0	3.2	2.5	2.5	1.7	0.64				1.2	2.7
n	5	5	5	5	5	5				5	5
s	0.5	0.9	0.7	0.4	0.6	0.14				0.2	0.9
Median	2.2	2.9	2.1	2.5	1.4	0.69				1.1	2.9
Flounder muscle tissue											
Average	3.4	3.1	3.1	3.2	2.5	1.9	3.8	3.3	1.8	2.5	1.7
n	25	20	25	25	25	25	25	25	20	25	25
s	0.8	1.7	1.8	1.2	1.1	1.2	2.4	2.7	0.7	1.8	2.0
Median	3.3	2.4	2.4	2.9	2.0	1.7	3.1	2.1	1.8	2.0	0.9
Flounder liver											
Average	4.9	3.3	4.4	3.6	3.3	2.5	4.6	4.9	1.7	2.8	1.8
n	5	5	5	5	5	5	5	5	4	5	4
s	1.8	1.6	0.8	0.45	1.2	1.3	1.9	2.6	0.5	0.6	0.6
Median	5.8	2.3	4.4	3.9	2.8	2.0	4.8	3.9	1.7	2.7	2.0
Blue mussel											
Average	2.5	2.3	1.90	2.3		0.85	3.7	1.7	1.6	1.3	1.6
n	4	4	4	4		4	4	4	4	4	4
s	0.3	0.2	0.03	0.4		0.04	0.5	0.1	0.2	0.2	0.1
Median	2.4	2.3	1.91	2.2		0.84	3.6	1.6	1.6	1.3	1.6
Brown shrimp											
Average		0.50	0.49	0.71	0.35	0.28	0.49	0.21	0.17	0.29	0.26
n		13	5	5	5	5	5	2	5	5	5
s		0.08	0.09	0.21	0.06	0.07	0.18	0.02	0.02	0.03	0.05
Median		0.49	0.47	0.72	0.33	0.26	0.47	0.21	0.18	0.29	0.28

content (gravimetrically determined). The authors concluded that this kind of normalisation may provide a basis for extrapolation of PCB pollution data among species. However, the inter-tissue and inter-species variability of our contaminant data is on the same order of magnitude as that observed by Delbeke *et al.* [13]. Consequently, there may be some doubt whether speciation of the lipids would give an improvement in this case.

Length and sex had no noticeable effects on the PCB concentrations expressed on a fat weight basis, with one exception: PCB concentrations in cod liver significantly increased with length. An influence of the length of cod on the PCB content in the liver was previously reported by de Boer [14], who demonstrated a significant concentration difference between individuals of different sizes (53-54 cm and 85-91 cm). Similarly, Kruse and Krüger [15] measured higher DDT levels in liver of Baltic cod of larger size, but they did not notice similar trends for hexachlorobenzene (HCB), α-hexachlorocyclohexane (α-HCH) or dieldrin. Bioaccumulation of contaminants such as PCBs in biota is the result of a combination of uptake (directly from the water, ingestion of contaminated particles and food) and elimination (metabolisation, excretion, growth dilution, spawning). The relative importance of each process will, of course, depend on the species considered and its stage of life. An explanation for the size-dependent contaminant level in cod liver may be found in the regime of larger cod. Larger cod mainly feeds on fish, which is more contaminated than invertebrates that are preferably consumed by smaller fish [16]. The major route of PCB uptake in larger cod, food, will therefore cause biomagnification. The bioaccumulation in muscle tissue is, however, not size-dependent; this may be related to the fact that lipid deposition with cod is mainly in the liver. The food consumption pattern of flounder, mussel and shrimp does not change during their life cycle [17]. For those species, no significant biomagnification was found; obviously, the uptake of PCBs is compensated by elimination processes.

Significant downward trends were observed in muscle tissue of cod and flounder, and in shrimp, but not in mussel and the liver of both fish species. From among these species, flounder, blue mussel and brown shrimp are excellent indicator organisms which clearly reflect the quality status of their habitats because of no or restricted migratory activities. Cod has a more enhanced migratory behaviour and does not necessarily reflect the condition of the area of capture. Nevertheless, cod is considered to be a suitable biomonitor for spatial and temporal trend monitoring. Migration appears to be sufficiently confined and allows observing differences between regions that are some hundred kilometres apart [18]. The observed temporal trends in this study are on the same tenor as others recently reported. In the 1993 North Sea Quality Status Report [2], decreasing PCB contents were cited for several species and various locations were cited and recent observations revealed decreasing concentrations of lower-chlorinated PCBs in yellow eel (*Anguilla anguilla*) from inland waters in the Netherlands [19]. PCB concentrations in cod (*Gadus morhua*) from the North Sea have been shown to have decreased significantly, although higher chlorinated congeners remained at an essentially constant level [19]. Constant PCB contents were reported by Stronkhorst [20] for

Mytilus edulis and by Solé *et al.* [21] for *Mytilus galloprovincialis* from the western Mediterranean. The observed trends may well indicate that PCB concentrations have reached their maximum values and that the compulsory remedial actions implemented by national and international organisations to improve the quality of the marine environment gradually become successful. However, although PCB concentrations are shown to decrease regionally, a global decline should not be expected in the next few years, because of on-going inputs into the environment caused by e.g. leakages from landfills and emissions from incinerators [22]. Moreover, it has been stated that the quantities of PCBs still in use, still exceed the amount that has been released into the environment to date [22].

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