SCOTTISH ENVIRONMENT PROTECTION AGENCY

Internal Draft Report:

Occurrence and Significance of Polychlorinated Biphenyls (PCBs) in Marine Sediments From Scottish Fish Farm Locations

D.Pirie

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Distribution: David MacKay, Andy Rosie, Brian Clelland, Calum McPhail

Summary

PCBs and organic carbon were determined in sediments from the vicinity of Scottish Fish Farms. The highest concentrations were detected close to fish farm cages suggesting that the farms were the source of the PCBs. Comparison of PCB concentrations with sediment quality standards indicates that the majority of sites are classified as slightly contaminated. The concentrations of PCBs recorded at the majority of sites are unlikely to cause biological effects.

Introduction

Recent press and TV reports have suggested that salmon feed contains significant concentrations of polychlorinated biphenyls (PCBs). There is concern that these persistent bio-accumulative chemicals may be accumulating in sediments around Scottish fish farms. A number of archived sediment samples from farms around the west coast of Scotland were analysed for PCBs. This report outlines the results of these analysis and compares the determined concentrations of PCBs with those reported in sediments from within the Clyde sea area, other UK marine sediments and marine sediments from around the world. The environmental significance of the concentrations is assessed by the use of marine sediment quality guidelines.

Background

Polychlorinated biphenyls (PCBs) are complex mixtures of chlorine substituted biphenyls. They are prepared by reacting biphenyl with chlorine in the presence of a catalyst such as iron (III) chloride. The resultant product is not a pure compound but a mixture of congeners with differing degrees of chlorination (Tanabe, 1988). These mixtures were marketed under trade names such as Aroclor, Phenoclor and Clophen. One of the largest manufacturers of PCBs was Monsanto (Law, 1993). Their nomenclature typically designates the degree of chlorination, for example Aroclor 1242 as the fraction containing 42 % of chlorine, Aroclor 1254 as 54 % chlorine and Aroclor 1268 as 68 % chlorine. PCB formulations have been prepared industrially since 1929, the highest production was during the sixties and early seventies. Environmental concerns led to severe restrictions on their sale and manufacture in most industrialised countries in the mid to late seventies, however, production continued in some countries such as the former Soviet Union until the early nineties (Colburn, 1996). Within the UK, PCB production commenced in 1954 and ceased in 1977, within that period a total of 66,748 tonnes were produced (Harrad *et al.*, 1994).

PCBs are exceptionally stable non flammable compounds highly resistant to oxidation, acids, bases, and other strong chemical reagents. In addition, they are excellent electrical insulators (Tatsukawa & Tanabe, 1987). These characteristics have resulted in PCBs being used in a variety of industrial applications. Prior to 1970, PCBs were used primarily in closed or semi-closed systems within electrical transformers, capacitors, heat transfer systems, and hydraulic fluids. They were also used to a minor extent in paints ,adhesives, caulking compounds, plasticisers, inks, lubricants, carbonless copy paper, sealants, coatings, and dust control agents.

PCBs have been shown to be ubiquitous environmental contaminants with their residues detected in all parts of the globe (Bommanna *et al.*, 1994). There are theoretically 209 possible PCB congeners and it is not analytically possible to accurately quantify all of the individual congeners present in environmental samples (Duinker *et al.*, 1988). To avoid confusion as to which congeners should be quantified and allow comparison of results from different research groups the International Council For the Exploration of the Seas (ICES) have recommended that seven congeners (with chlorine numbers from 3 -7) should be monitored in environmental samples. These so called ICES 7 congeners are listed below:

Table 1 ICES 7 PCB Congeners.

Congener No.	Chemical Structure
28	2,4,4' trichlorobiphenyl
52	2,2',5,5' tetrachlorobiphenyl
101	2,2',4,5,5' pentachlorobiphenyl
118	2,3',4,4',5 pentachlorobiphenyl
138	2,2',3,4,4',5' hexachlorobiphenyl
153	2,2',4,4',5,5' hexachlorobiphenyl
180	2,2',3,4,4',5,5' heptachlorobiphenyl

Throughout this report total PCBs have been estimated as the sum of the ICES 7 congeners multiplied by 2.5.

Location

Details of sampling locations with hydrographic conditions and an indication of farm biomass are presented in Table 2. Samples were collected at the cage edge and where possible at a reference site at least 500m from the cages using a 0.025m^2 van Veen grab. Sediment was sub-sampled through top opening flaps on the grab and stored in high density polyethylene pots. The sediment was then frozen aboard the survey vessel prior to transportation to the laboratory.

Table 2 Sampling Location Details

Description	Date	Grid Reference	Hydrographic Conditions	Consented Biomass (tonnes)	Max Biomass 1999	Max Biomass 2000 *
Meall Mhor Station 14	7/6/99	NR 186554 674268	moderately flushed	900	797	259
Cairndow Reference 3	4/6/99	NN 210181 597834	poorly flushed	700	0	0
Cairndow Station 4	4/6/99	NN 212187 599317				
Rubha Stillaig Station 18	8/6/99	NR 192536 668388	moderately flushed	900	938	601
Rubha Stillaig Reference 8	8/6/99	NR 192444 668885				
Furnace Quarry Station 7	5/6/99	NN 203655 700043	poorly flushed	450	453	430
Lamlash Bay Reference 10	9/6/99	NR 203927 629807	moderately flushed	1250	1172	540
Lamlash Bay Station 21	9/6/99	NR 204359 629271				
Lamlash Bay Reference 13	31/7/00	NS 203927 629807				
Lamlash Bay Station 17	31/7/00	NR 204215 629361				
Craig Lodge Station 19	1/8/00	NS 200430 677563	poorly flushed	500	363	218
Ardchattan Station 16	26/7/00	NM 197251 734705	poorly flushed	140	To find out	to find out
Port na Moine Station 2	18/7/00	NM 180533 701426	poorly flushed	500	453	484
Loch Spelve B Station 6	20/7/00	NM 168966 726979	poorly flushed	700	653	382
Loch Spelve Reference 3	20/7/00	NM 168521 727510				
Charlotte Bay Station 8	21/7/00	NM 183379 730641	moderately flushed	600	986	254
Port na Moralachd Station 11	22/7/00	NM 181985 745328	moderately flushed	1000	0	437
Baig an Spairbh Station 20	1/8/00	NS 209340 669463	moderately flushed	1000	991	937

* Year 2000 max biomass figures are to September 2000 only.

Where poorly flushed is defined as mean current speed less than 5cm/s, moderately flushed is defined as mean current speed 5-10 cm/s and well flushed is defined as mean current speed greater than 10cm/s.

Chemical Analysis

The sediment samples were frozen, freeze dried, then gently sieved through a nylon 500μm mesh to produce a free flowing powder. The dried samples were stored at room temperature in sealed high density polyethylene pots for up to a year prior to analysis. The stability over time of dried sediments stored in this manner has not been rigorously tested, however, dried sediment used as a laboratory reference material has been stored under similar conditions for up to 3 years without significant change within our laboratory.

Sub samples of the sediments were analysed for total nitrogen and organic carbon using a CE instruments NA2500 nitrogen and carbon analyser. The samples were completely oxidised at 1000°C and the combustion components separated and detected by a thermal conductivity detector.

A portion of the powder was soxhlet extracted using acetone (33%) and hexane (67%). Sulphur containing compounds were removed by reaction with elemental copper during the extraction. The resulting extract was reduced in volume using tubovap concentration. Fats, lipid and other co-extracted materials were removed by adsorption chromatography using sulphuric acid silica and silver-nitrate alumina columns.

PCBs were separated, identified and quantified by gas chromatography with electron capture detection. A mid polarity HP Ultra 2 column of 50M length, 0.2mm inner diameter and 0.33μm film thickness was used to perform the separation. Compounds eluting from the GC column were identified by comparison of their retention times to those of certified standards of individual PCB congeners. Quantification was by comparison of peak heights with multipoint calibration graphs of certified standards and internal standards.

Prior to routine use this analytical procedure was subjected to rigorous in house validation according to NS30 (Cheeseman *et al.*, 1989). The routine precision and accuracy of the procedure was maintained by the analysis of a procedural blank, a recovery standard, a laboratory reference material and a duplicate sample with each batch of samples. In addition the accuracy of this method has been validated by participation in inter laboratory comparison exercises such as the National Marine Monitoring Program AQC scheme, and the Quasimeme European quality assurance programme. Our laboratory is UKAS accredited for the determination of PCBs in sediments.

Results

A total of 18 sediments from Scottish farmed salmon sites were analysed for PCBs and organic carbon by SEPA West laboratory based at East Kilbride. The results are presented below:

Table 3 Concentration of PCBs and Organic Carbon in Sediments

		Concentration µg/kg Dry Weight									%	Estimated Total PCB
	Date of Sampling	PCB 28	PCB 52	PCB 101	PCB 118	PCB 153	PCB 138	PCB 180	Sum ICES 7	Estimated Total PCB	Organic Carbon	Normalised to 1% Organic Carbon
Meall Mhor Sataion 14	06/07/99	<0.86	0.79	1.59	1.32	3.02	<0.05	0.89	8.52	21.30	2.67	7.98
Cairndow Reference 3	06/04/99	<1.41	<0.14	0.40	0.42	0.82	<0.08	<0.08	3.35	8.37	0.56	14.9
Cairndow Station 4	06/04/99	<1.46	<0.14	<0.22	<0.17	0.17	<0.08	<0.08	2.31	5.78	8.99	0.64
Rubha Stillaig Station 18	06/08/99	<1.40	0.45	0.84	0.66	1.43	<0.08	0.38	5.23	13.07	7.29	1.79
Rubha Stillaig Reference 8	06/08/99	<0.98	<0.09	<0.08	<0.06	0.15	<0.05	<0.06	1.47	3.69	0.62	5.95
Furnace Quarry Station 7	06/05/99	<1.42	0.47	0.67	0.60	1.27	<0.08	0.53	5.04	12.60	4.71	2.68
Lamlash Bay Reference 10	06/09/99	<1.30	<0.13	<0.10	<0.08	0.13	<0.07	<0.07	1.87	4.70	0.92	5.1
Lamalsh Bay Station 21	06/09/99	<1.44	1.45	2.20	1.92	4.14	3.95	1.13	16.23	40.57	18.52	2.19
Lamlash Bay Reference 13	31/7/00	<0.13	<0.14	0.29	0.25	0.45	0.47	<0.07	1.80	4.50	1.24	3.63
Lamlash Bay Station 17	31/7/00	<0.17	<0.17	0.34	0.25	0.52	0.54	<0.09	2.08	5.20	3.42	1.52
Craig Lodge Station 19	08/01/00	1.24	1.10	1.45	0.81	1.88	1.85	0.58	8.91	22.27	4.39	5.07
Ardchattan Station 16	26/7/00	1.26	1.20	1.56	0.97	1.86	1.81	0.51	9.16	22.90	6.85	3.34
Port na Moine Station 2	18/7/00	<0.15	<0.16	<0.12	<0.09	0.46	0.44	<0.08	1.50	3.76	4.57	0.82
Loch Spelve B Station 6	20/7/00	0.56	0.71	1.32	1.14	2.30	2.25	0.66	8.94	22.36	7.69	2.91
Loch Spelve Reference 3	20/7/00	<0.16	<0.16	<0.12	<0.10	<0.10	<0.08	<0.09	<0.81	2.02	3.66	0.55
Charlotte Bay Station 8	21/7/00	0.42	<0.16	0.65	0.61	1.30	1.23	0.32	4.69	11.74	7.29	1.61
Port na Moralachd Station 11	22/7/00	<0.16	<0.16	<0.12	0.22	0.26	0.29	<0.09	1.31	3.28	3.87	0.85
Baigh an Spairbh Station 20	08/01/00	1.22	0.88	1.27	1.22	2.11	2.24	1.16	10.10	25.26	4.71	5.36

Concentrations of PCBs detected in the sediments were in the range 2-40µg/kg dry weight, and organic carbon was in the range 0.5 -15%. In general, the highest concentrations of PCBs and organic carbon were detected at the sites closest to the cages. PCB concentrations significantly elevated above reference sites were detected at the following locations; Lamlash Bay Station 21, Loch Spelve Station 6 and Rubha Stillaig Station 18. This suggests that fish farms are a source of PCBs and that PCB residues are accumulating in the vicinity of Fish Farms.

Comparison with Clyde Sea Sediments

The following tables list the concentrations of PCBs detected in sediments from surveys conducted by SEPA West in the Clyde Sea area (Note these are not normalised to 1% organic carbon).

Table 4 Estimated Total PCB concentrations in Sediments from the Clyde

Location	UK Grid Reference	Date of Sampling	Estimated Total PCBs µg/kg Dry Weight
Clyde at Elvanfoot	NS 957 184	25/11/93	2.7
Clyde at Bonnington Weir	NS 885 407	25/11/93	3.4
Clyde at Motherwell Bridge	NS 737 563	25/11/93	3.0
Clyde at B758 road Bridge	NS 685 617	25/11/93	4.2
Clyde at Cambuslang Bridge	NS 641 610	25/11/93	4.8
Clyde at Dalmarnock Bridge	NS 617 627	25/11/93	68.3
Clyde at RutherGlen Bridge	NS 605 630	25/11/93	51.4
Clyde at Tidal Weir	NS 595 644	25/11/93	76.7
Clyde Estuary up stream of Shieldhall STW	NA	02/02/ 93	91.5
Clyde Estuary down stream of Shieldahall STW	NA	02/02/ 93	82.7
Clyde Estuary up stream Dalmuir STW	NA	02/02/ 93	111.2
Clyde Estuary down stream Dalmuir STW	NA	02/02/93	67.7
Clyde Estuary Control Site	NA	02/02/93	118.4

Table 5 Estimated Total PCB concentrations in Sediments from the Cart Estuary

Location	UK Grid Reference	Date	Total PCBs µg/kg Dry Weight
Cart Estuary up stream Paisley STW	NA	03/02/93	136.8
Cart Estuary down stream Paisley STW	NA	03/02/93	96.6
Cart Estuary Control Site	NA	03/02/93	540.3
Cart Estuary Babcocks Harbour	NA	03/02/93	215.1
Cart Estuary down stream Babcocks Harbour	NA	03/02/93	566.8
Cart Estuary White Cart Bridge	NA	03/02/93	112.3
Cart Estuary Colins Isle	NA	03/02/93	42.6

NA - Not Available

The majority of sediments from the fish farm sites contained less than 20 µg/kg total PCBs. This is significantly lower than the concentration of PCBs detected in sediments from the lower reaches of the Clyde and Cart Estuaries. The sediments in which slightly elevated PCBs concentrations were detected contained concentrations comparable to those found in sediments from the lower reaches of the Clyde and Cart Estuaries.

Comparison with Other Marine Sediments

The following table lists concentrations of PCBs reported in marine sediments from around the UK.

Table 6 Summary of Estimated Total PCBs Concentrations in UK Marine Sediments.

Location	Year	%TOC	Total PCB Concentration µg/kg dry Weight	Reference
Inner Firth of Clyde	1989	3.3 - 4	40-112.5	Kelly *et al* 1995
Firth of Clyde - Incmarnock Water	1989	1.4 - 4.2	12.8 - 35	
Mid/South Firth of Clyde	1989	0.9 - 3.1	4.5 - 17.2	
Outer Firth of Clyde	1989	0.5 - 1.4	1.2 - 16.7	
Garroch Head Former Sludge Dump Site	1989	1.1 - 5.1	102 - 1250	
Garroch Head Sludge Dump Site	1989	2.4 - 11.9	25 - 305	
Liverpool Bay	1988	ND	13.7 - 37.9	Camacho-Ibar *et al* 1996
Shandon Dock (Liverpool)	1988	ND	320	
Inner Thames Estuary	1993	ND	3 - 40	Scrimshaw *et al* 1995
Tyne Estuary	1990	ND	6.1 - 81	MAFF 1991
Humber	1990	ND	3 - 7.6	
Cardigan Bay	1990	ND	0.2 - 3.9	
Morecambe Bay	1990	ND	0.2 - 4.7	
Humber, Tees, Tweed and Ribble Estuaries	1990	ND	0.2 - 20	ENDS 1993
Tyne, Wear and Mersey Estuaries	1990	ND	20 -100	
North Sea	1986	0.1 - 15	0.1 - 4.4	Knickmeyer *et al* 1988
North Sea	1986-1987	0.4 14.8	0.21 - 9.18	Knickmeyer *et al* 1990

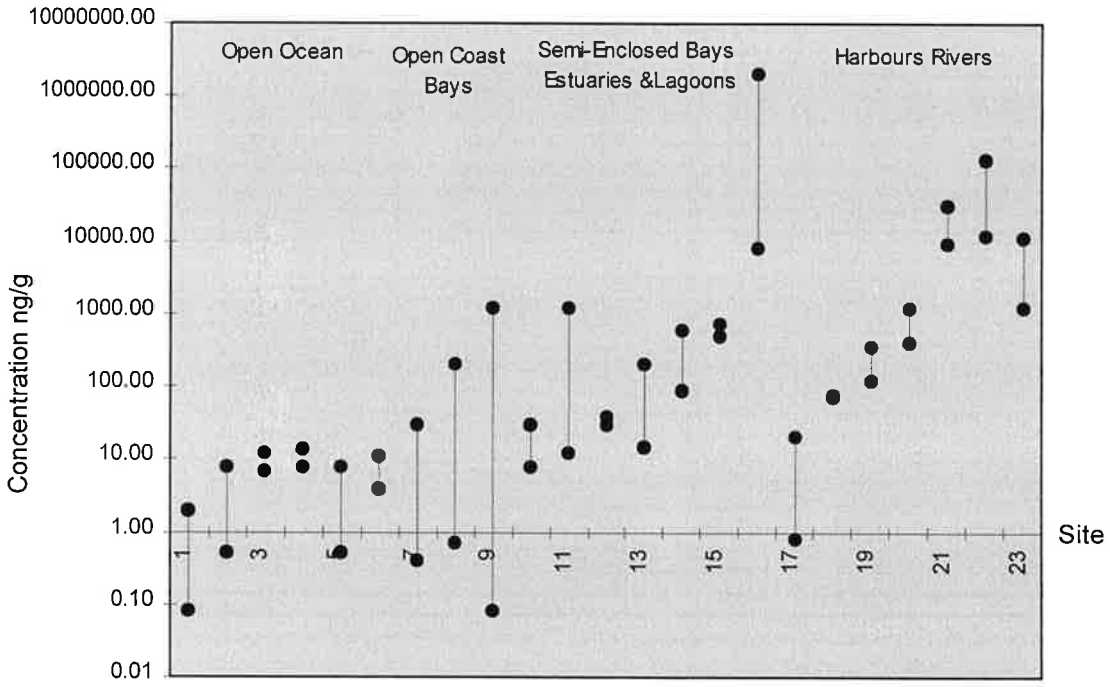
ND - Not Determined

The concentrations of PCBs detected in the sediments from around fish farms are similar to concentrations reported in marine sediments from open locations around the UK but are lower than the concentrations reported for industrialised estuaries e.g. Clyde, Tyne, Wear, Mersey and Thames estuaries.

Typical background concentrations of Total PCB in soils from around the UK are 2 - 32 µg/kg, typical concentrations in sewage sludge are between 106 and 712 µg/kg (DOE 1994). Total PCB concentrations at the majority of the sites are similar to background concentrations in soil from around the UK and are significantly lower than those recorded in sewage sludge from around England and Wales.

Total PCB concentrations in marine sediments from around the world are shown visually in Figure 1. This shows a clear concentration gradient with the lowest concentrations recorded in the open oceans and the highest concentrations reported in harbours. The concentrations reported in the sediments are in the mid to lower reaches of this scale. This is typical for open coast and semi-enclosed bays and confirms that the concentrations of PCBs in the fish farm sediments are many orders of magnitude lower than highly contaminated sites.

Figure1 Estimated Total PCB concentration ranges in sediments from around the world.



Where: 1 = North Sea (Lohse, 1988); 2 = North sea (Boon *et al.*, 1985); 3 = Humber Plume (Klamer & Foomsgaard, 1989); 4 = Biscay Bay (Grimalt *et al.*, 1992); 5 = Thermaikos Gulf (Larsen & Fytianos, 1986); 6 = Hano Bight (Larsson, 1984); 7 = Liverpool Bay (Camacho-Ibar & McEvoy, 1996); 8 = Casco Bay (Kennicutt *et al.*, 1994), 9 = Gulf of Mexico Coast (Sericano *et al.*, 1990), 10 and 11 Tolo Bay and Junk Bay (Kannan *et al.*, 1989); 12 = Lagoon of Venice (Pavoni *et al.*, 1987), 13 = River Elbe Estuary (Japenga *et al.*, 1987); 14 = San Diego Bay (McCain *et al.*, 1992); 15 = Buzzards Bay (Beller & Simoneit, 1986); 16 = New Bedford Harbour Estuary (Pruell *et al.*, 1990); 17 = Manukau Harbour (Fox *et al.*, 1988); 18 = Long Beach Harbour (Beller & Simoneit, 1986); 19 = Rotterdam Harbour (Japenga *et al.*, 1987); 20 and 21 = New Bedford Harbour (Brownawell & Farrinton, 1986); 22 = Gothenburg Harbour (Jarnberg *et al.*, 1993); Upper Hudson River (Bush *et al.*, 1987).

Comparison with Quality Standards

Currently the UK has not set marine sediment quality standards for PCBs. However, a number of marine monitoring groups have recognised the need for a method of classifying sediment contaminant concentrations and have suggested guideline concentrations for contaminants in marine sediments and these are summarised in Table 7.

I. Following the Piper - Alpha oil platform incident in the North Sea SOAEFD defined a series of concentration guidelines in order to categorise the levels of chlorinated biphenyls in marine sediments (Wells *et al.*, 1989). These guidelines have been used to categorise sediments from around the UK coast (MAFF,1991) and have become widely accepted as an appropriate method of classifying PCB concentrations in marine sediments. Under these guidelines sediments in the range 0.2-20μg/kg are classed as "slightly contaminated", sediments in the range 21-100μg/kg are classed as "contaminated" and those >100μg/kg as highly contaminated".

II. The Florida Department of Environmental Protection (FDEP) have developed effects based sediment quality assessment guidelines (SQAG) for 34 priority substances in Florida coastal waters (Florida Department of Environmental Protection, 1994). These guidelines have been developed using the weight of evidence approach. Using this approach a data base is compiled containing information generated by the three approaches to the establishment of effects based sediment quality guidelines: the equilibrium approach, the spiked sediment toxicity approach, and the evaluation of matching sediment chemistry and biological effects data. The data in the database is weighted and used to compile the sediment quality assessment guidelines. These quality guidelines are designed for the evaluation of marine sediments around Florida, caution must be exercised when comparing these guidelines to marine sediments from colder northern waters. The guidelines suggest that the threshold level for effects is 21.6 μg/kg and the probable effects level is 189 μg/kg.

III. Using similar methods the Joint Monitoring Group of the Oslo and Paris Commissions have also derived from studies which contained biological effects studies (Fleming *et al* ., 1995), standard sediment concentrations at below which biological effects are unlikely to occur. The majority of studies were North American/Canadian and as previously indicated the values should be used with caution. The guideline indicates that biological effects are unlikely at the 1-10 μg/kg level.

These guideline sediment assessment concentrations are summarised below in Table6. The SOAEFD/MAFF guidelines do not account for sediment bound organic carbon, the FDEP and JMP guidelines are both for 1%TOC normalised sediments.

Table 7 Summary of Marine Sediment Quality Assessment Guidelines.

	SOAEFD / MAFF	F.D.E. P		JMG
Compound	Sediment Categories ng/g dry weight	Threshold effects level (TEL) μg/kg dry weight	Probable effects level (PEL) μg/kg dry weight	Sediment concentrations below which biological effects unlikely to occur. μg/kg dry weight normalised to 1% organic carbon
PCBs	<0.2 contamination not detectable 0.2 - 20 slightly contaminated 21 -100contaminated >100 heavily contaminated	21.6	189	1-10

The majority of the sediments monitored in this study from the vicinity of fish farms in the West Coast of Scotland are classified as slightly contaminated using the SOAEFD/MAFF sediment classification scheme. Six sites would be classified as contaminated. The concentrations of total PCBs recorded in sediments from all of the sites were lower than the FDEP threshold effects level and the majority of sites were below the JMG concentration at which biological effects were unlikely to occur.

Conclusions

PCBs were detected in the vicinity of Scottish fish farms. The highest concentrations were detected close to the cages suggesting that the farms were the source of the PCBs. The concentrations of total PCBs reported in sediments are the similar to those reported for open coastal and semi enclosed lagoons within the UK and Europe but slightly higher than pristine Scottish coastal waters . Comparison with sediment quality standards indicates that the majority of sites are classified as slightly contaminated. The concentrations of PCBs recorded at all the sites are below FDEP threshold effects concentrations and unlikely to cause biological effects at the majority of sites.

Future Work

PCBs are persistent bioaccumulative materials with half lives measured in decades. This very limited initial study has suggested that PCBs may be accumulating close to fish farm sites. It is suggested that further detailed spatial and depth studies be undertaken at high, medium, and low dispersion sites. These should consist of sediment cores collected in a grid around representative farms. Sediment cores will allow an estimation of the historical inputs of PCBs to be made. Full detailed congener analysis should be undertaken to allow evaluation of different sources e.g. atmospheric versus source point inputs.

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