

# Levels of Organochlorine Compounds, Including PCDDs and PCDFs, in the Blubber of Cetaceans from the West Coast of North America

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Levels of organochlorine compounds (PCDD, PCDF, PCB and organochlorine pesticides) were determined in cetaceans collected from the west coast of North America between 1986 and 1989. The samples included gray whale (*Eschrichtius robustus*), killer whale (*Orcinus orca*), false killer whale (*Pseudorca crassidens*), Risso's dolphin (*Grampus griseus*) and Dall's porpoise (*Phocoenoides dalli*) collected in British Columbia, and harbour porpoises (*Phocoena phocoena*) collected in British Columbia and central California. TCDD and TCDF levels ranged from 1 to 8 ng kg<sup>-1</sup> and 2.0 to 109 ng kg<sup>-1</sup>, respectively. The highest levels of PCDDs were found in the harbour porpoises; the levels of 1,2,3,6,7,8-HxCDD in the samples from Victoria, Campbell River and Qualicum River were 128, 128 and 62 ng kg<sup>-1</sup>, respectively. Five other 2,3,7,8-substituted dioxins and dibenzofurans were detected in the cetaceans at levels ranging from 1 to 10 ng kg<sup>-1</sup>. In addition to the 2,3,7,8-substituted congeners, several non 2,3,7,8-substituted congeners were detected. The patterns of the PCDDs and PCDFs in the British Columbia porpoises were consistent with implication of chlorophenols as the source of the PCDDs and PCDFs, which were either present in wood chips used in bleached kraft paper mills, or came from direct contamination by chlorophenols. No PCDDs or PCDFs were detected in the California samples. One false killer whale sample had exceptionally high levels of DDT compounds (1700 mg kg<sup>-1</sup> DDE, 120 mg kg<sup>-1</sup> DDT and 40 mg kg<sup>-1</sup> *o,p'*-DDT) and Toxaphene (89 mg kg<sup>-1</sup>). PCB levels in the cetaceans were highest in the false and killer whales (22 to 46 mg kg<sup>-1</sup> GM), and lowest in the Risso's dolphin (1.7 mg kg<sup>-1</sup>). Levels of DDE in the British Columbia harbour porpoises were 6.0 mg kg<sup>-1</sup>, and probably reflect the accumulation of global background levels of DDE.

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persistent organochlorine compounds to levels far above ambient ocean concentrations (Jensen *et al.*, 1969; Addison *et al.*, 1984; Muir *et al.*, 1988). Some of the first reports of organochlorine contamination in the marine environment were reported in cetaceans (Holden & Marsden, 1967; Gaskin *et al.*, 1971). This, together with the observation of both possible reproductive impairment and population declines in marine mammals, has led to many studies over the last two decades on the levels and effects of contaminants on cetaceans (O'Shea & Brownell, 1994; Tanabe *et al.*, 1994). Despite the relatively large amount of data concerning contaminants in cetaceans (especially in harbour porpoises, *Phocoena phocoena*), there is very little recent data from the west coast of North America.

In a study of organochlorine contaminants in the blubber of cetaceans from the Pacific and south Atlantic Oceans, O'Shea *et al.* (1980) analysed five species from the coast of California collected between 1968 and 1976 for DDT compounds, PCBs, dieldrin, HCB, toxaphene and chlordane compounds. In this study, the highest levels of PCBs and DDT compounds were found in California samples (O'Shea *et al.*, 1980). Calambokidis *et al.* (1984) reported the levels of PCBs and DDE in the blubber of five species of cetaceans collected from Washington State and British Columbia between 1976 and 1981. They reported high levels of DDE and PCB in two killer whale (*Orcinus orca*) samples (640 mg kg<sup>-1</sup> DDE and 250 mg kg<sup>-1</sup> PCB, and 59 mg kg<sup>-1</sup> DDE and 38 mg kg<sup>-1</sup> PCB) and in one harbour porpoise (14 mg kg<sup>-1</sup> DDE and 55 mg kg<sup>-1</sup> PCB); and low concentrations (below 10 mg kg<sup>-1</sup>) in one Dall's porpoise (*Phocoenoides dalli*; Calambokidis *et al.*, 1984). Varanasi *et al.* (1994) analysed organochlorine and metal contaminants in a variety of tissues (and stomach contents) of 22 grey whales (*Eschrichtius robustus*) found stranded along the west coast of North America. The levels of ΣPCBs (total of the PCB congeners) and ΣDDTs in blubber ranged from 0.12 to 10 and 0.009 to

Most marine mammals (apart from baleen whales) are high trophic level predators, and bioaccumulate

2.1 mg kg<sup>-1</sup>, respectively; no differences were found in contaminant levels between regions (Varanasi *et al.*, 1994).

Beginning in 1987, a volunteer programme was initiated by the Marine Mammal Research Group (MMRG) in British Columbia to monitor the number of cetacean strandings and incidental catches, the species involved, and the geographical and seasonable distribution of such events (Guenther *et al.*, 1992). Other goals of this group are to maximize the research and educational use of dead animals through necropsies and to collect samples for toxicology, parasitology, genetic analysis, and for life history and feeding habit studies (Guenther *et al.*, 1992).

Research into polychlorinated dibenzo-*p*-dioxin (PCDD) and polychlorinated dibenzofuran (PCDF) contamination in British Columbia began as a result of a survey of PCDD contamination in fish-eating birds in Canada (Norstrom & Simon, 1983). High levels of PCDDs (up to 740 ng kg<sup>-1</sup> 1,2,3,6,7,8-hexachlorodibenzo-*p*-dioxin, 1,2,3,6,7,8-HxCDD) were discovered in the pooled eggs of the great blue heron (*Ardea herodias*) from a colony on the Endowment Lands of the University of British Columbia (UBC; Elliott *et al.*, 1989). Since the herons are year-round residents in coastal British Columbia, the PCDD and PCDF contamination was presumed to be of local origin. This monitoring programme was extended to include three more colonies of herons in the Strait of Georgia in 1983. Two of these sites were found to be more contaminated than the UBC site (Elliott *et al.*, 1989). Based on the profiles of the PCDDs and PCDFs congeners, the source of the contamination was proposed to be bleached kraft paper mills, and chlorophenols, which are used as a wood fungicide (Elliott *et al.*, 1989).

The goal of this study was to determine the levels of organochlorine contaminants in the blubber of a variety

of cetacean species collected by the MMRG in British Columbia, mainly in the Strait of Georgia and the Pacific coast of Vancouver Island, and to compare these samples to samples collected in California. The toxicological significance of these levels, the potential sources of the contaminants, and a comparison of these levels to those reported for other cetacean populations are discussed.

## Materials and Methods

### Sample collection

The blubber samples used in this study were collected by the MMRG and the California Marine Mammal Stranding Program. The specimen number, collection location, age, sex and date collected are presented in Table 1 and Fig. 1. Collection details of British Columbia specimens have been presented by Langelier *et al.* (1990).

### Chemical analysis

*PCDDs and PCDFs (Canadian Wildlife Service)*. A complete description of the method can be found in Norstrom & Simon (1991). Briefly, the method was as follows: the samples were blended with 100 ml of hexane for 10 min in a Sorval Omni-Mixer three times (20–25 g of anhydrous Na<sub>2</sub>SO<sub>4</sub> was added following the first extraction), and filtered through a layer of Na<sub>2</sub>SO<sub>4</sub>. All of the filtrates were combined, the volume was reduced and then brought back to 24–48 ml hexane:dichloromethane (1:1). At this stage the sample was spiked with a cocktail of five <sup>13</sup>C labelled tetra- to octa-chloro PCDD congeners, which were used as recovery standards. In order to remove the bulk of the lipid, the samples were cleaned up by automated gel-permeation chromatography (GPC). A portion of the extract was sealed in glass ampules for determination of organochlorines and PCBs by the Department of

TABLE 1  
Blubber sample collection locations, age, sex and date collected.

	Specimen no.	Collection location	Age	Sex	Date collected
<i>Eschrichtius robustus</i>	SWDP 87-15	Long Beach, V.I.	Calf	U	9/87
	SWDP 88-05	Denman Island	Calf	M	5/88
<i>Orcinus orca</i>	SWDP 86-05	Port Renfrew, V.I.	Mature	F	U
	SWDP 86-04	Tsawwassen	Neonate	M	7/86
	SWDP 87-16	Ucluelet, V.I.	Calf	M	11/87
	SWDP 89-02	Radar Beach, V.I.	17 years	M	4/89
	SWDP 89-12	Namu	8 years	M	6/89
	SWDP 89-X	Stuart Island, WA	New Born	M	1/89
<i>Pseudorca crassidens</i>	SWDP 87-06	Denman Island	Mature	M	3/387
	SWDP 89-21	Vargas Island, V.I.	Mature (old)	M	9/89
<i>Grampus griseus</i>	SWDP 88-03	Queen Charlotte Island	Mature	M	3/88
<i>Phocoenoides dalli</i>	SWDP 87-09	Sooke, V.I.	Immature	F	5/87
	SWDP 88-07	White Rock	Mature	M	5/88
<i>Phocoena phocoena</i>	SWDP 88-04	Saanich Penn., V.I.	Immature	M	5/88
	SWDP 87-07	Campbell River, V.I.	Mature	F	5/87
	SWDP 87-12	Gabriola Island	Calf	F	8/87
	SWDP 87-17	Sandspit, Moresby Island	Imm/Mat	M	11/87
	SWDP 87-18	Long Beach, V.I.	Imm/Mat	M	12/87
	SWDP 87-19	Tsawwassen	Imm/Mat	M	12/87
	SWDP 88-06	Victoria, V.I.	Imm/Mat	M	5/88
	SWDP 89-09	Qualicum Beach, V.I.	Mature	F	5/89
	LML 87-9	Monterey Bay, CA	Mature	F	10/87
	LML 88-1	Monterey Bay, CA	Immature	M	10/87
LML 88-2	Monterey Bay, CA	Immature	F	3/88	

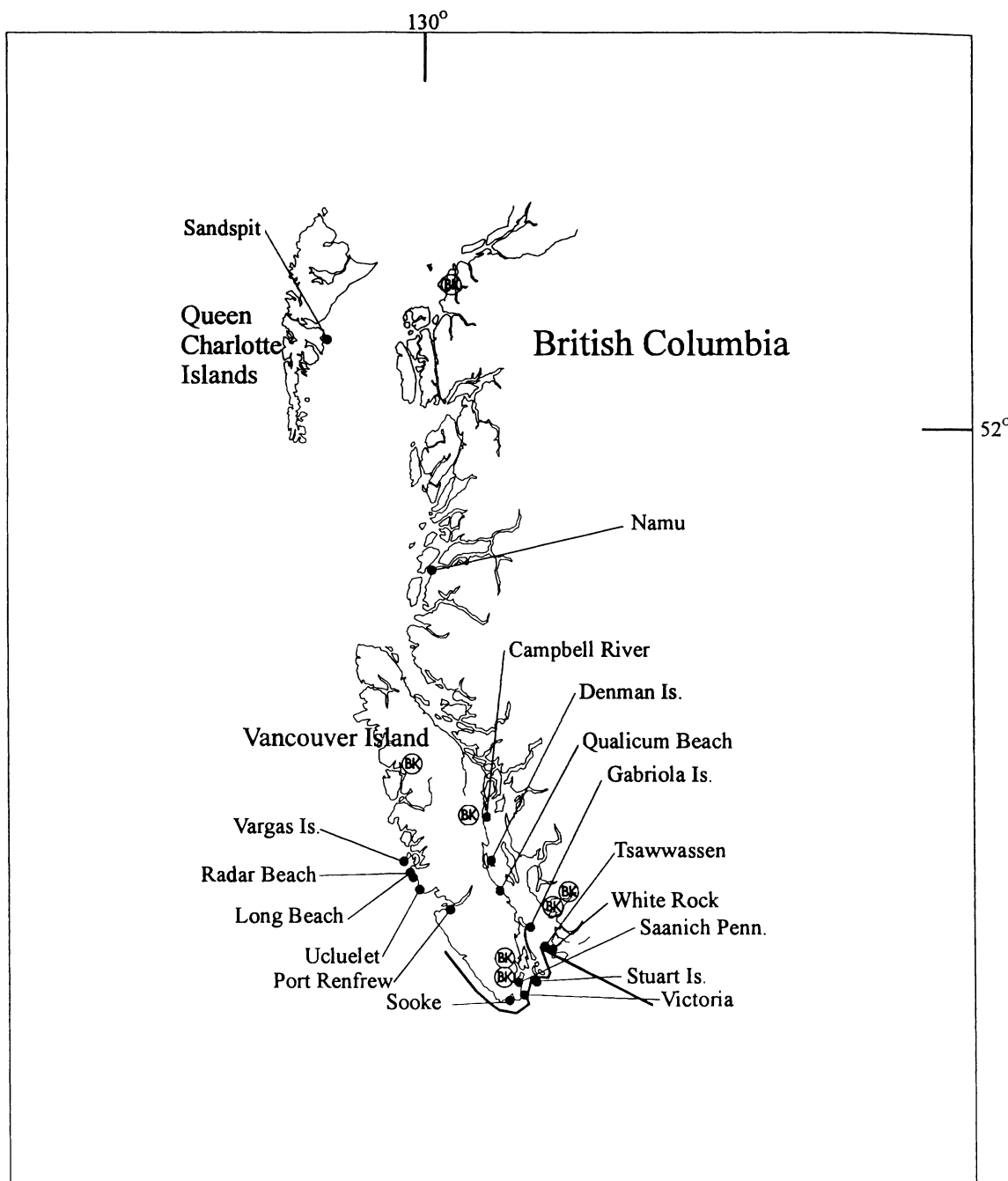


Fig. 1 Sample collection sites (except the California samples). BK is the approximate location of bleached kraft paper mills.

Fisheries and Oceans. The extracts were further cleaned up by running the extract from the GPC through a carbon column/glass fibre column and eluting the PCDDs and PCDFs from the carbon column in reverse direction. The samples were spiked just prior to instrumental analysis with  $^{37}\text{Cl}$ -2,3,7,8-TCDD which was used as a performance internal standard. The samples were analysed on a Hewlett Packard (HP) 5987B GC/MS, operated in selected ion mode (SIM), using a  $30\text{ m} \times 0.25\text{ mm}$  i.d. DB-5 column (J&W Scientific). Quantitation was performed using the internal standard method.

*Organochlorines and PCBs (Department of Fisheries and Oceans)*. A complete description of the method can be found in Muir *et al.* (1988) and Ford *et al.* (1993). Briefly, the method is as follows: sub-samples were

mixed with  $\text{Na}_2\text{SO}_4$  and extracted with hexane using a small ball mill.  $^{13}\text{C}$ -PCBs 77, 126 and 169 were added at the initial extraction and were used as recovery and quantitation standards. Lipid was determined gravimetrically on 1/100 of the sample. Lipid removal was performed using an automated GPC. Ten percent of the sample was removed for organochlorine and ortho PCB analysis. The remaining extract was eluted on a silica gel column, and passed through a carbon/glass fibre column. The non-ortho PCBs were reverse eluted off the carbon column. Instrumental analysis was performed on a HP GC/MS mass selective detector operated in SIM mode, using a  $30\text{ m} \times 0.25\text{ mm}$  i.d. DB-5 column (J&W Scientific). Non-ortho PCBs were quantified using the internal standard method. Organochlorines and ortho PCBs were separated into three

fractions on a Florisil column according to the procedure described by Norstrom *et al.* (1988a). Instrumental analysis was performed using a Varian 6000 GC equipped with a <sup>63</sup>Ni ECD, and a 60 m × 0.25 mm i.d. DB-5 column (J&W Scientific) using hydrogen carrier gas. PCBs and OC pesticides were quantified as described in Muir *et al.* (1988) using external standards.

**Results and Discussion**

*Isomer specific polychlorinated dibenzo-p-dioxin and polychlorinated dibenzofuran analysis*

2,3,7,8-tetrachloro dibenzo-*p*-dioxin (TCDD) was detected in four of the harbour porpoises (1–5 ng kg<sup>-1</sup>), one killer whale sample (2 ng kg<sup>-1</sup>), and one false killer whale sample (8 ng kg<sup>-1</sup>; Table 2). Buckland *et al.* (1990) reported levels of 4 to 11 ng kg<sup>-1</sup> TCDD in Hector's dolphins from New Zealand, and Beck *et al.* (1990) reported levels of TCDD in the harbour porpoise from the North Sea at less than 0.5 ng kg<sup>-1</sup>. Trace levels (1–13 ng kg<sup>-1</sup>) of 1,2,3,7,8-pentachloro-*p*-dioxin (1,2,3,7,8-PnCDD), 1,2,3,7,8,9-hexachloro-*p*-dioxin, 1,2,3,4,6,7,9-heptachloro-*p*-dioxin, 1,2,3,4,6,7,8-heptachloro-*p*-dioxin and octachloro-*p*-dioxin were also detected in the harbour porpoise samples from British Columbia, killer whale, and false killer whale samples (Table 2). No dioxins were detected in the California harbour porpoise, Risso's dolphin or grey whale samples (Table 2).

The most commonly detected PCDD or PCDF congener was 2,3,7,8-tetrachloro dibenzofuran (TCDF). The highest levels of TCDF were detected in one of the false killer whales (109 ng kg<sup>-1</sup>), and in the Dall's porpoises (16–69 ng kg<sup>-1</sup>; Table 2). TCDF was the only PCDF congener detected in the Risso's dolphin or grey whales (Table 2). Levels of TCDF in killer whales were 6–43 ng kg<sup>-1</sup>. No other PCDFs were detected in any of the whales. The observation of relatively more PCDDs than PCDFs in killer whales and false killer whales is opposite to that for killer whales collected off the Pacific coast of Japan (Ono *et al.*, 1987). It was reported by Ono *et al.* (1987) that there were much higher PCDF levels (300–480 ng kg<sup>-1</sup> total PCDFs) than PCDD levels (not detectable at <0.3–10 ng kg<sup>-1</sup>) in the killer whales, suggesting from the congener profiles that these high levels were associated with PCDFs found in PCBs (high levels of PCBs were also found in the whales 350–410 mg kg<sup>-1</sup>).

The highest levels of PCDDs were found in the harbour porpoises from British Columbia (Table 2); the levels of 1,2,3,6,7,8-hexachloro dibenzo-*p*-dioxin (1,2,3,6,7,8-HxCDD) in the samples from Victoria, Cambell River and Qualicum River were 128, 128 and 62 ng kg<sup>-1</sup>, respectively. These levels are more than an order of magnitude higher than those reported for Hector's dolphin (*Cealorhynchus hectori*) from New Zealand (Buckland *et al.*, 1990), or for levels of PCDDs in a harbour porpoise from the North Sea (Beck *et al.*, 1990). The highest level of 1,2,3,6,7,8-HxCDD detected in the killer whales was 14 ng kg<sup>-1</sup> in the calf from Tsawwassen and 23 ng kg<sup>-1</sup> in the false killer whale from Denman Island (Table 2).

**TABLE 2**

Levels of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans in cetacean blubber from the west coast of North America, collected between 1986 and 1989. Levels reported as ng kg<sup>-1</sup> of the wet weight.

Specimen no.	Species																							
	Grey Whale			Killer Whale			False Killer Whale			Risso's Dolphin		Dall's Porpoise			Harbour Porpoise (BC)			Harbour Porpoise (CA)						
	88-5	87-15	86-2	86-1	87-16	89-2	89-12	89-X	87-6	89-21	88-3	88-4	88-7	87-09	88-6	87-7	87-12	87-17	87-18	89-9	87-19	1 ml 87-9	1 ml 88-1	1 ml 88-2
2,3,7,8-TCDD	<2	<2	2	<2	<2	<2	<2	2	8	<2	<2	<2	4	2	1	2	<2	<2	<2	4	5	<2	<2	<2
1,2,3,7,8-PnCDD	<5	<5	8	<5	5	5	1	5	8	1	<5	<5	9	4	4	19	<5	<5	<5	9	<5	<5	<5	<5
1,2,3,6,7,8-HxCDD	<8	<8	14	<8	2	<8	7	9	23	2	<8	5	22	10	128	128	<8	<8	<8	62	<8	<8	<8	<8
1,2,3,7,8,9-HxCDD	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	1	2	10	10	<8	<8	<8	6	<8	<8	<8	<8
1,2,3,4,6,7,9-HpCDD	<10	<10	<10	<10	2	<10	<10	<10	<10	<10	<10	<10	3	5	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
1,2,3,4,6,7,8-HpCDD	<10	<10	<10	<10	3	<10	2	<10	<10	<10	<10	2	4	5	8	8	<10	<10	<10	6	<10	<10	<10	<10
OCDD	<20	<20	<20	<20	<20	<20	5	<20	<20	<20	<20	10	16	15	<20	<20	<20	<20	<20	8	<20	<20	<20	<20
2,3,7,8-TCDF	3	<2	39	23	13	13	6	43	109	2	4	16	69	21	11	43	11	2	4	38	31	<2	<2	<2
1,2,4,7,8-PnCDF	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	10	16	10	<5	5	<5	<5	<5	<5	<5	<5	<5	<5
2,3,4,7,8-PnCDF	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	4	<5	<5	<5	<5	<5	<5	<5	<5
1,2,4,8,9-PnCDF	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	5	9	4	<5	4	<5	<5	<5	<5	<5	<5	<5	<5
1,2,4,6,8,9-HxCDF	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	8	12	<8	<8	10	<8	<8	<8	<8	<8	<8	<8	<8
1,2,3,4,6,9- <i>l</i>	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	4	3	<8	<8	5	<8	<8	<8	<8	<8	<8	<8	<8
1,2,3,6,8,9-HxCDF	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	6	13	<10	<10	12	<10	<10	<10	<10	<10	<10	<10	<10
1,2,3,4,6,8,9-HpCDF	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	6	13	<10	<10	12	<10	<10	<10	<10	<10	<10	<10	<10

In 1992, B.C. Environment reported the levels on 2,3,7,8-substituted PCDD and PCDF, and several organochlorine compounds in seven harbour porpoises and one killer whale found stranded in the Strait of Georgia (Burlinson, 1991). Of the PCDDs and PCDFs only 2,3,7,8-TCDD and TCDF were reported. TCDD was detected in only one of the harbour porpoises ( $3.3 \text{ ng kg}^{-1}$ ) and in the killer whale ( $2.3 \text{ ng kg}^{-1}$ ); and TCDF ranged from not detected to  $68 \text{ ng kg}^{-1}$  (Burlinson, 1991). These levels are similar to those reported in this study (Table 2).

Of particular interest was the detection of non 2,3,7,8-substituted PCDF congeners in the Dall's porpoises and harbour porpoises from British Columbia. The major bioaccumulating PCDD and PCDF compounds in mammals have been reported to be the 2,3,7,8-substituted congeners (Abraham *et al.*, 1989; van den Berg & Poiger, 1989). Reports of PCDDs and PCDFs in dolphins and porpoises are limited, but none report non 2,3,7,8-substituted congeners (Beck *et al.*, 1990; Buckland *et al.*, 1990). As mentioned above, Ono *et al.* (1987) reported a variety of non 2,3,7,8-substituted congeners in killer whales that also were highly contaminated with PCBs from the Pacific coast of Japan, and attributed these congeners to PCDFs in commercial PCBs; therefore, it may be that cetaceans lack the ability to readily metabolize PCDD and PCDF congeners, as normally metabolized by mammals (Tanabe *et al.*, 1988). Conversely, TCDD may be metabolized more readily by cetaceans than other mammals (Norstrom *et al.*, 1992).

The major environmental sources of PCDDs and PCDFs are various kinds of combustion, contaminants in pesticides, bleached kraft paper mills and PCBs (PCDFs only; Rappe, 1992). Bleach Kraft pulp and paper mills produce TCDD and greater amounts of TCDF (Amendola *et al.*, 1989). High levels of TCDD and TCDF have been reported in crabs from waters east of Vashon Island (Strait of Georgia) and these levels have been attributed to the effluent of bleached kraft paper mills (Norstrom *et al.*, 1988b). The widespread occurrence of TCDD and TCDF in the cetacean samples from British Columbia, and their virtual absence from the California harbour porpoise samples, indicates the local influence of the British Columbia pulp mills.

A variety of PCDD and PCDF congeners have been reported in both tetrachlorophenol and pentachlorophenol (PCP; Rappe *et al.*, 1978; Miles *et al.*, 1985; Hagenmaier & Brunner, 1987), and PCDFs have been reported in commercial PCBs (Wakimoto *et al.*, 1988). The major PCDD congener (1,2,3,6,7,8-HxCDD) in the porpoise samples from British Columbia is not found in large amounts in atmospheric (or combustion) samples (Rappe *et al.*, 1989), but is found in commercial chlorophenols (Rappe *et al.*, 1978). In addition to 1,2,3,6,7,8-HxCDD, 1,2,3,7,8-PnCDD is also found in herons and crabs as well as the cetaceans from British Columbia (Elliott *et al.*, 1989; Norstrom *et al.*, 1988b). Characteristic PCDFs that were found in chlorophenols (Rappe *et al.*, 1978; Hagenmaier & Brunner, 1987), and in the same porpoise samples from British Columbia,

include 1,2,4,7,8-pentachlorodibenzofuran, 1,2,4,8,9-pentachlorodibenzofuran, 1,2,4,6,8,9-hexachlorodibenzofuran and 1,2,3,4,6,8,9-heptachlorodibenzofuran. The dominance of 1,2,3,6,7,8-HxCDD among PCDDs in all British Columbia cetaceans pattern is consistent with the HxCDD source being pulp mills, using PCP-contaminated feed stock. Formation of 1,2,3,6,7,8-HxCDD, and probably 1,2,3,7,8-pentachloro dibenzo-*p*-dioxin, occurs by intermolecular condensation of phenoxyphenol impurities in the PCP during the pulping process (Luthe *et al.*, 1992).

One prominent PCDF congener in chlorophenols (1,2,4,6,8-pentachloro dibenzofuran), and one prominent PCDD congener (1,2,3,6,7,9/12,3,6,8,9-hexachloro dibenzo-*p*-dioxin) which are both present in chlorophenols (Rappe *et al.*, 1978; Hagenmaier & Brunner, 1987) and in crab hepatopancreas from British Columbia (Norstrom *et al.*, 1988b), were not detected in the dolphins. However, the general patterns of co-occurrence of these PCDD and PCDF congeners in both chlorophenols and dolphins implicate chlorophenols, either present in wood chips used in bleached kraft paper mills, or direct contamination by chlorophenols, as the source of PCDDs and PCDFs in the dolphins.

Because cetaceans have been shown to accumulate non 2,3,7,8-substituted PCDFs, the lack of non 2,3,7,8-substituted PCDF congeners in the whales from British Columbia is probably related to diet, rather than metabolism. Fish are able to metabolize non-2,3,7,8 PCDF congeners (Sijm *et al.*, 1993), although measurable amounts may be found in fish under high levels exposure situations. In the Strait of Georgia, killer whales tend to be generally piscivorous, or they tend to consume a large number of pinnipeds (Baird *et al.*, 1992). Porpoises consume a variety of invertebrates (Recchia & Read, 1988), which do not have the same metabolic capacity as fish (Rappe & Buser, 1989). Norstrom *et al.* (1988b) found a variety of non 2,3,7,8-substituted furans in crab hepatopancreas from the Strait of Georgia. It is possible that the non 2,3,7,8-substituted dibenzofurans detected in the porpoises is a reflection of their consumption of invertebrates, in combination with their limited metabolic capacity.

#### *Organochlorine Pesticide and PCBs*

In general, the highest levels of organochlorine compounds were found in the false killer and killer whale samples (the grey whale samples were not analysed for OCs and PCBs) (Table 3). DDE levels for the Denman Island false killer whale sample have been previously reported (Baird *et al.*, 1989). The false killer whale collected on Vargas Island also had elevated DDE, toxaphene and PCB levels of 60, 17 and  $46 \text{ mg kg}^{-1}$ , respectively.

The levels of DDE and PCBs in the killer whales (28 and  $22 \text{ mg kg}^{-1}$  geometric mean (GM), respectively) and the toxaphene levels are much lower than in the false killer whales ( $6.2 \text{ mg kg}^{-1}$  GM vs 89 and  $17 \text{ mg kg}^{-1}$ ). The levels are similar to those reported by Calambokidis *et al.* (1984) for one resident killer whale ( $59 \text{ mg kg}^{-1}$  DDE and  $38 \text{ mg kg}^{-1}$  PCB). As mentioned above, resident killer whales in British

TABLE 3

Organochlorine levels in cetacean blubber from the west coast of North America, collected between 1986 and 1989. Concentrations reported as  $\mu\text{g kg}^{-1}$  of the wet weight with geometric mean (GM), one standard deviation (SD), and number of samples in which the residues were detected (N).

Number of samples analysed % Lipid (Average)	Killer Whale			False Killer Whale			Risso's Dolphin			Dall's Porpoise			Harbour Porpoise			California									
	Killer Whale			False Killer Whale			Risso's Dolphin			Dall's Porpoise			Harbour Porpoise			California									
	GM	GM + / -SD	N	89-21	93	87-6	1	30	3	98	GM	GM + / -SD	N	92	GM	GM + / -SD	N	73	GM	GM + / -SD	N				
28 000	(9600-84 000)	6	60 000	1 700 000	4200	4300	(1100-17 000)	3	6000	(2100-17 000)	7	12 000	(9400-15 000)	3	1100	(490-2500)	7	1400	(1100-1700)	3	3	1400	(1100-1700)	3	
1900	(770-4400)	6	2300	35 000	110	520	(120-2200)	3	1100	(490-2500)	7	1400	(1100-1700)	3	490	(110-2200)	7	1300	(1100-1600)	3	3	1300	(1100-1600)	3	
1300	(640-2600)	6	7100	120 000	340	540	(170-1700)	3	490	(110-2200)	7	1400	(1100-1700)	3	190	(60-610)	7	240	(180-330)	3	3	240	(180-330)	3	
580	(200-1700)	6	6600	40 000	370	84	(17-420)	3	190	(60-610)	7	240	(180-330)	3	8200	(3100-22 000)	7	15000	(12 000-18 000)	3	3	15000	(12 000-18 000)	3	
32 000	(11 000-93 000)	6	76 000	1 900 000	5000	5500	(1400-21 000)	3	8200	(3100-22 000)	7	15000	(12 000-18 000)	3	28		1	9.8			1	9.8		1	
<1		0	<1		<1		<1	0	<1		0	<1		0		0	<1			0	<1		0		
3.3	(2.3-4.8)	3	1	16	<1	4.6	(1.6-13)	2	11	(3.6-36)	4	<1		11		4	<1			4	<1		0		
450	(140-1500)	6	520	110	33	140	(13-1500)	3	470	(240-950)	7	350	(150-810)	3	250	(170-350)	7	240	(120-470)	3	3	240	(120-470)	3	
77	(33-180)	6	100	58	11	130	(13-1300)	3	250	(170-350)	7	240	(120-470)	3	800	(370-1700)	7	500	(300-830)	3	3	500	(300-830)	3	
600	(320-1100)	6	640	90	34	1400	(920-2300)	2	800	(370-1700)	7	500	(300-830)	3	84	(45-150)	7	60	(29-130)	3	3	60	(29-130)	3	
31	(17-58)	6	43	12	4.1	110	(70-190)	2	9.2	(7.4-11)	2	20	(17-23)	2	34	(13-88)	2	85	(31-230)	2	2	85	(31-230)	2	
<1		0	1	5.5	<1	18	(8-40)	2	<1		0	<1		7.5		1	12	(7-21)		1	12	(7-21)	1		
<1		0	1	1	1	<1	(16-19)	2	17	(16-19)	2	17	(16-19)	2	<1		0	6.6	(2.6-17)		0	6.6	(2.6-17)	0	
<1		0	25	13	13	7.5	(2.5-22)	2	<1		0	<1		25	(7.7-84)	6	18	(9.8-32)		6	18	(9.8-32)	6		
42	(14-130)	6	73	93	<1	23	(14-38)	2	23	(14-38)	2	23	(14-38)	2	8	(2.6-25)	6	7.4		6	7.4		1		
18	(4.8-69)	6	40	130	2.9	16	(11-24)	2	16	(11-24)	2	16	(11-24)	2	<1		0	0.98		0	0.98		1		
17		1	1	1	<1	<1		0	<1		0	<1		<1		0	0			0		0	1		
280	(110-730)	6	470	310	26	110	(16-750)	3	250	(110-580)	7	200	(77-510)	3	180	(77-410)	7	85	(55-130)	3	3	85	(55-130)	3	
120	(41-370)	6	110	98	8.8	190	(140-270)	3	180	(77-410)	7	200	(77-510)	3	32	(12-85)	7	28	(17-46)	3	3	28	(17-46)	3	
34	(8.4-130)	6	110	1800	11	42	(33-52)	2	42	(33-52)	2	42	(33-52)	2	340	(140-820)	7	250	(140-430)	3	3	250	(140-430)	3	
320	(110-950)	6	640	330	41	110	(18-660)	3	340	(140-820)	7	250	(140-430)	3	2500	(1000-5800)	7	1600	(660-3900)	3	3	1600	(660-3900)	3	
3000	(1100-8200)	6	5400	3800	310	900	(120-6900)	3	900	(120-6900)	3	900	(120-6900)	3	300	(140-660)	7	300	(140-670)	3	3	300	(140-670)	3	
300	(130-680)	6	400	190	19	130	(19-880)	3	300	(140-660)	7	300	(140-670)	3	3700	(1600-8400)	7	2600	(1100-5800)	3	3	2600	(1100-5800)	3	
4200	(1600-11 000)	6	7200	6800	420	1800	(340-9000)	3	1800	(340-9000)	3	1800	(340-9000)	3	520	(230-1200)	7	84	(7.7-920)	3	3	84	(7.7-920)	3	
340	(150-770)	6	280	360	20	670	(460-970)	3	670	(460-970)	3	670	(460-970)	3	15	(3.6-8.1)	7	9.2	(1.6-53)	2	2	9.2	(1.6-53)	2	
12	(3.7-12)	5	1	1	<1	17	(11-26)	2	17	(11-26)	2	17	(11-26)	2	5.4	(3.6-8.1)	7	21	(9.3-48)	3	3	21	(9.3-48)	3	
54	(30-97)	6	170	310	6.4	32	(7.8-130)	3	32	(7.8-130)	3	32	(7.8-130)	3	3800	(1500-9700)	7	2200	(1100-4600)	3	3	2200	(1100-4600)	3	
6200	(1500-25 000)	6	17 000	89 000	750	1300	(120-14 000)	3	1300	(120-14 000)	3	1300	(120-14 000)	3			6	N.A.		6	N.A.		6		
1.9	(1.6-2.3)	6	0.59	3.4	1.6	1.7	(0.94-3.1)	3	1.7	(0.94-3.1)	3	1.7	(0.94-3.1)	3	1.7	(0.95-3)	6	N.A.		6	N.A.		6		
1	(0.52-1.9)	6	0.31	1	1	1.6	(0.46-5.7)	3	1.6	(0.46-5.7)	3	1.6	(0.46-5.7)	3	0.65	(0.29-1.5)	7	N.A.		7	N.A.		7		
1.1	(0.42-2.7)	6	0.62	4.5	0.39	1.9	(1.3-2.9)	3	0.71	(0.34-1.5)	7	N.A.		0.71	(0.34-1.5)	7	N.A.		7	N.A.		7	N.A.		
3.7	(2-6.6)	6	1.3	5.5	0.76	3.7	(0.94-15)	3	2.8	(1.3-6)	7	N.A.		2.8	(1.3-6)	7	N.A.		7	N.A.		7	N.A.		
169	(0.4-0.63)	5	1.5	2.8	<0.1	0.6	(0.4-0.91)	3	0.6	(0.19-1.9)	4	N.A.		0.6	(0.19-1.9)	4	N.A.		4	N.A.		4	N.A.		
22 000	(8600-56 000)	6	46 000	34 000	1700	4500	(470-43 000)	3	8400	(4400-16 000)	7	10 000	(3300-31 000)	3			7	10 000		7	10 000		7	10 000	

N.A., Not Analysed.

\*False killer whales presented as individual data.

†Labelling of chlordanes compounds according to Dearth & Hites (1991).

Columbia water feed mainly on fish, as opposed to transitory killer whales whose diet consists of a large proportion of pinnipeds (Baird *et al.*, 1992).

Ono *et al.* (1987) reported levels of  $\Sigma$ PCBs ranging from 350 to 410 mg kg<sup>-1</sup> in killer whale blubber from the coast of Japan, and Kemper *et al.* (1994) reported average levels of 28.4 mg kg<sup>-1</sup> in killer whales from Australia. Although the levels of PCBs reported here for the false killer and killer whales appear relatively high (> 20 mg kg<sup>-1</sup>), they appear to be at the lower range of concentrations reported for killer whales.

Profiles for PCB congeners representing greater than 1% of the total PCBs are similar between the killer whale and false killer whales from Vargas Island; however the false killer whales from Denman Island has a different congener profile (Fig. 2). The major congeners in the killer whale and false killer whales from Vargas Island were CB 153 > CB 138 > CB 149  $\approx$  CB 99  $\approx$  CB 118  $\approx$  CB 101. The major congeners in the false killer whales from Denman Island were PCB 153 > PCB 180  $\approx$  PCB 84/89 > PCB 187 > PCB 138. Muir *et al.* (1988) reported very little alteration of PCB profiles between cetaceans and Arctic cod; therefore these different patterns probably reflect a different feeding strategy or sources among the individual whales.

Both the killer whales and false killer whales are long-lived high trophic level predators, and in the case of the false killer whales, both samples were collected from mature males, factors which could additionally contribute to the accumulation of OC contaminants (Subramanian *et al.*, 1987). Despite these factors, the high levels of OCs in the false killer whales are difficult to explain fully. There are no other reported levels of organochlorine compounds in false killer whales for

comparison. In a survey of contaminants in cetaceans collected from the Pacific and Atlantic Oceans, O'Shea *et al.* (1980) found the highest levels of DDE in southern California samples (as high as 2500 mg kg<sup>-1</sup> DDE in bottlenose dolphin blubber). They attributed these high levels to DDT originating from a manufacturing plant in Los Angeles (MacGregor, 1974). The distribution of false killer whales is primarily tropical and sub-tropical (Baird *et al.*, 1989); it is possible the elevated levels of DDE in the false killer whale were accumulated in southern California. However, O'Shea *et al.* (1980) found no detectable levels (> 0.1 mg kg<sup>-1</sup>) of toxaphene in their California samples. Therefore, if the DDE is assumed to come from the Southern California Bight, the accumulation of high levels of toxaphene by the false killer whales has to be separate from the accumulation of DDE, perhaps in Central America where toxaphene is still heavily used (Voldner & Li, 1993).

The highest levels of *p,p'*-DDE found in the dolphins and porpoises were in the California harbour porpoises (12 mg kg<sup>-1</sup> GM). Levels of DDE in the British Columbia harbour porpoise samples were approximately 6.0 mg kg<sup>-1</sup>, and the levels of DDE in the Dall's porpoise and Risso's dolphin were similar (4.3 and 4.2 mg kg<sup>-1</sup>, respectively). Both Risso's dolphin and Dall's porpoise are in general a more deep water or offshore species than harbour porpoises, whose habitat is coastal and offshore shallows (Jefferson, 1990; Baird & Stacey, 1991; Gaskin, 1992). The similar and low values for DDE in these three species probably reflects the bioaccumulation of background levels of DDE in the Pacific ocean.

Profiles for PCB congeners representing greater than 1% of the total for the dolphins and porpoises are presented in Fig. 3. The harbour porpoises from British Columbia and California have very similar profiles to those in killer whales, with the relative contribution of congeners being CB 153 > CB 138 > CB 149. The Risso's dolphin and Dall's porpoises profiles have a greater predominance of the lesser chlorinated congeners (i.e. < 6 chlorines). Notably, the most abundant congeners in Dall's porpoise were CB 118 > CB 152 > CB 153 > CB 183. The Risso's dolphin and Dall's porpoise are more of an open water or open ocean species as opposed to the harbour porpoises which inhabit coastal areas. This difference of PCB patterns between inshore and offshore cetacean species was also noted by Duinker *et al.* (1989) and may be related to diet differences (e.g. fish vs squid).

Recently, high levels of PCBs have been associated with striped dolphin (*Stenella coeruleoalba*) mortalities in the Mediterranean (Aguilar & Borrell, 1994). The mean levels of  $\Sigma$ PCBs in the blubber of the striped dolphins was 393 mg kg<sup>-1</sup> (wet wt; Kannan *et al.*, 1993b). This is more than 30 times the highest levels found in the dolphins and porpoises in this study. Kannan *et al.* (1993b) found a high ratio of PCB 169 to PCB 126 in the highly contaminated dolphins, and found a good linear correlation between this ratio (PCB 169/PCB 126) and the  $\Sigma$ PCB concentration. They suggested that, because of the high levels of PCBs, there was an increase of the metabolic activity of the

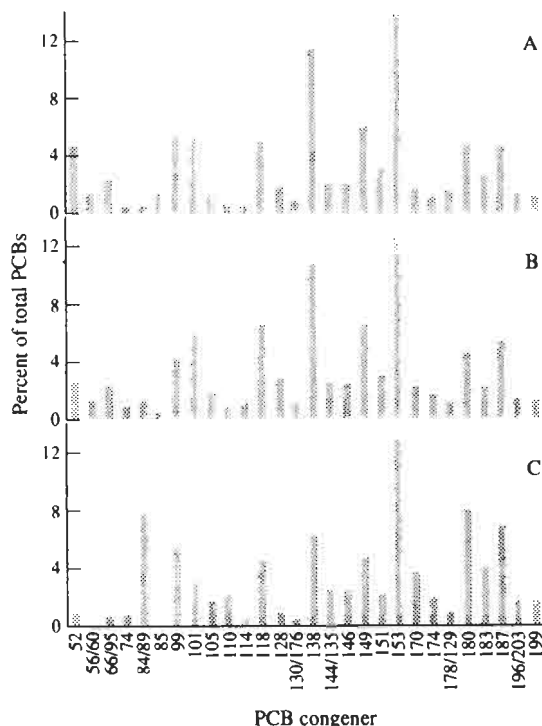


Fig. 2 PCB congener profiles for congeners that constitute greater than one percent of the total mixture. A, Killer whale geometric mean; B, false killer whale, Vargas Island; C, false killer whale, Denman Island.

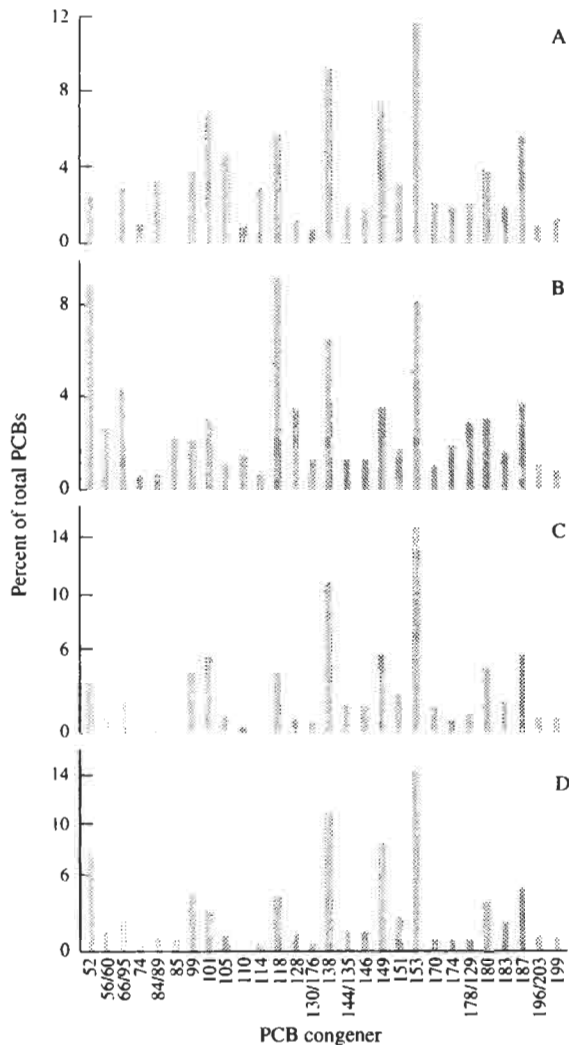


Fig. 3 PCB congener profiles for congeners that constitute greater than 1% of the total mixture. A, Risso's dolphin; B, Dall's porpoise; C, harbour porpoise, California; D, harbour porpoise, British Columbia.

mixed function oxidase system (in particular CYP450 1A), resulting in the relative degradation of PCB 126. Furthermore, they suggested that the ratio of PCB 169 to PCB 126 could serve as an index for risk assessment in marine mammals. A regression of the ratio of PCB 169 to PCB 126 vs total PCB concentration for all samples in this study showed no statistical correlation ( $p > 0.001$ ). It is possible that this ratio is specific for PCB levels within a species, and may not be applicable across species, or that levels of PCBs were too low to cause induction of metabolic activity.

There is an extensive amount of literature reporting the levels of organochlorine contaminants in harbour porpoises (Holden & Marsden, 1967; Gaskin *et al.*, 1971; Koeman, 1972; Clausen *et al.*, 1974; Taruski *et al.*, 1975; Anderson & Rebsdorff, 1976; Kerkoff & de Boer, 1977; Harms *et al.*, 1978; O'Shea *et al.*, 1980; Gaskin *et al.*, 1983; Calambokidis *et al.*, 1984; Duinker *et al.*, 1988, 1989; Morris *et al.*, 1988; Beck *et al.*, 1990; Buckland *et al.*, 1990; Kannan *et al.*, 1993a; Kuiken *et al.*, 1994). In an attempt to identify temporal and regional trends in harbour porpoises, and how the values from this study relate to previously published values, the DDE concentrations

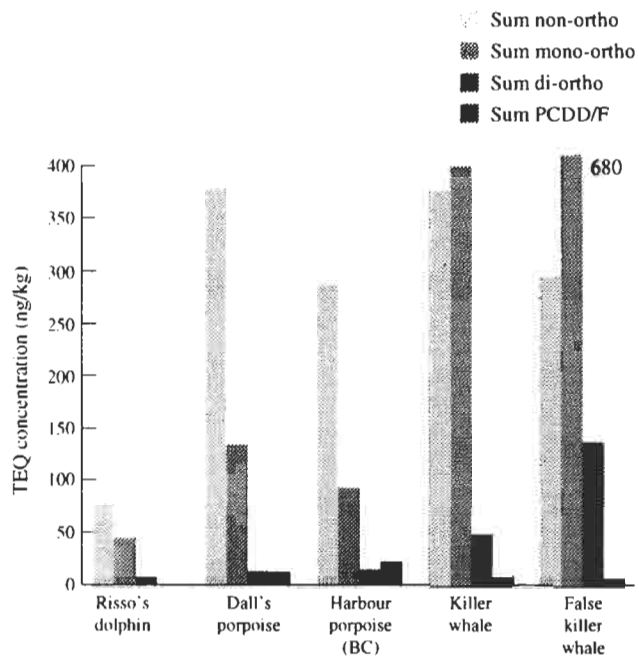


Fig. 4 TCDD equivalents for cetacean blubber from the west coast of North America. Concentration reported as  $\text{ng kg}^{-1}$  of the wet weight, geometric mean. The PCB TEFs were taken from Ahlborg *et al.* (1994), the PCDD and PCDF TEFs from Kutz *et al.* (1990).

were grouped into five regions and two time periods: the regions are California, the northeast Pacific, the northwestern Atlantic, the North Sea and the Baltic Sea; the time periods are 1965–1980 and 1981–1994 (Table 4). (Because of the difficulties of comparing temporal PCB levels generated by a variety of analytical methods, PCBs were not included in this data analysis.)

The levels of DDE in harbour porpoises from the northeast Pacific did not change dramatically over the two time periods. The relatively low levels of DDE in the northeastern Pacific and North Sea samples is additional evidence that these levels ( $< 10$  ppm) reflect the accumulation of global background levels of DDE. As mentioned earlier, O'Shea found high levels of DDE in southern California cetaceans, but the samples in the present study were collected in northern California waters (Monterey Bay), which is not highly contaminated with DDE (Phillips, 1988). It is possible that the slightly elevated levels of DDE in the recent California samples are a result of the continuing influence of the past dumping of DDT wastes in southern California. However, it is difficult to assess temporal trends in California harbour porpoises because of the different geographical sampling areas.

Historic levels of DDT in the northwest Atlantic harbour porpoises (primarily the Bay of Fundy) were higher than any other geographical area except California, but there is no recent information on the levels of DDE in harbour porpoises from this region. The levels of DDE in the North Sea are low ( $< 10$   $\text{mg kg}^{-1}$ ) and appear to be declining. Historic levels of DDE in porpoises from the Baltic sea is limited but appear to be higher than in the North Sea. Recently they have been shown to also be contaminated with high levels of PCBs (Kannan *et al.*, 1993a).



TABLE 4

DDE in harbour porpoise blubber grouped according to region and time period. Concentrations reported as mg kg<sup>-1</sup> of the wet weight, geometric mean (GM), and number of samples (N).

	NE Pacific*		California†		NW Atlantic‡		North Sea§		Baltic¶	
	GM	N	GM	N	GM	N	GM	N	GM	N
1965–1980	4.4	2	270	1	51	6	6.9	6	17	2
1981–1994	6.0	7	12	3	–	0	1.9	99	–	0

\*NE Pacific: 1965–1980 data from Calambokidis *et al.*, 1984; 1981–1994 data from this study.

†California: 1965–1980 data from O'Shea *et al.*, 1980; 1981–1994 data from this study.

‡NW Atlantic: 1965–1980 data from Gaskin *et al.*, 1971 (geometric mean of five averages of  $n = 12$ ,  $n = 2$ ,  $n = 1$ ,  $n = 15$  and  $n = 6$  samples), and Taruski *et al.*, 1975,  $n = 1$ ).

§North Sea: 1965–1980 data from Holden & Marsden, 1967 (geometric mean of one average of three samples and one sample), and Kerkoff & de Boer, 1977,  $n = 1$ ; 1981–1994 data from Beck *et al.*, 1990,  $n = 1$ , Morris *et al.*, 1989,  $n = 4$ , and Kuiken *et al.*, 1994,  $n = 94$ .

¶Baltic: 1965–1980 data from Harms *et al.*, 1978 (geometric mean of averages of two groups of five samples).

### TCDD equivalents

The 2,3,7,8-substituted dibenzo-*p*-dioxins and dibenzofurans and the 3,4,3',4'-substituted non- and mono-ortho substituted PCBs have similar chemical properties, and have been reported to share a common receptor-mediated mechanism of toxic action (Safe, 1990). Based on a number of *in vivo* and *in vitro* studies, a set of toxic equivalency factors (TEFs) have been proposed for these compounds, assigning the most toxic congener TCDD a value of one (all other TEFs are less than one; Safe, 1990; Ahlborg *et al.*, 1994). Multiplying the concentration of the compound by the TEF results in a TCDD equivalent (TEQ), which allows the estimation of the concentration of dioxin-like toxicity, and determination of the relative contributions of the PCDD, PCDF and PCB congeners to TCDD-like toxicity.

The highest total TEQs in this study were found in the false killer whale samples (Fig. 4) at 1100 ng kg<sup>-1</sup>. Total TEQs in killer whale blubber collected from the coast of Japan calculated from Kannan *et al.* (1988) were 6300 ng kg<sup>-1</sup>, approximately five times higher than the highest levels in this study. Kannan *et al.* (1993) reported total TEQs of 18 000 ng kg<sup>-1</sup> in striped dolphins affected by a morbillivirus in the western Mediterranean in 1990; the levels of TEQs in this study are well below the levels associated with the morbillivirus epizootic in the Mediterranean.

Kannan *et al.* (1988) reported that in Dall's porpoise, killer whale, striped dolphin and finless porpoise (*Neophocaena phocaenoides*) collected from the North Pacific, over 50% of the total TEQ was contributed by the mono-ortho PCBs. In this study the major contributor to the total TEQs was the non-ortho congeners in the dolphin and porpoise samples, and the mono-ortho PCBs in the whale samples (Fig. 4). The PCDDs and PCDF were minor contributors to the total TEQ in all species (Fig. 4). The relative contribution of non-ortho and mono-ortho PCBs varies in different commercial PCB mixtures (Kannan *et al.*, 1988); therefore the relative contribution of non- and mono-ortho PCBs in cetaceans is influenced both by source of the PCBs, and by selective metabolism and degradation (Tanabe *et al.*, 1988; Falandysz *et al.*, 1994).

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