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# Persistent Organic Pollutants (POPs) in British Columbia Harbour Seals and Killer Whales

R.F. Addison and P.S. Ross

Fisheries and Oceans Canada
Institute of Ocean Sciences
Sidney, B.C.
V8L 4B2

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**Abstract**

The Persistent Organic Pollutants (POPs) represent a wide range of environmental contaminants that have been introduced into the environment through a number of processes. The POPs include the insecticide DDT, the industrial polychlorinated biphenyls (PCBs), and the polychlorinated dibenzo-p-dioxins (PCDDs) and –dibenzofurans (PCDFs). Most POPs are lipophilic, resistant to breakdown, and bioaccumulate in the aquatic food chain, often reaching high concentrations in fish-eating biota. Toxic effects in laboratory animals, fish-eating animals and humans include reproductive impacts, immunotoxicity and neurotoxicity. Despite the implementation of regulations for several POPs over the past quarter century, many persist in environmental samples and continue to present a risk to the health of wildlife at the top of the food chain. POPs have been widely distributed around the world through atmospheric processes, compounding the influence of regional sources in British Columbia and adjacent waters. Recent evidence suggests that even low to moderate contaminant levels are affecting endocrine processes in BC harbour seals. The finding that BC's killer whales now represent some of the most contaminated marine mammals in the world underscores the need to better understand the sources and fate of such chemicals in the environment, as well as the health effects on high trophic level organisms. The use of marine mammals as "sentinels" provides an integrative measure of marine ecosystem contamination and the food chain upon which they depend.

**Résumé**

Les polluants organiques persistants (POP) regroupent un large éventail de contaminants qui ont été introduits dans l'environnement par un certain nombre de processus. Ces polluants comprennent l'insecticide DDT ainsi que les biphényles polychlorés (PCB) d'usage d'industriel, les polychlorodibenzo paradioxines (PCDD) et les polychlorodibenzo furanes (PCDF). La plupart des POP sont lipophiles, ils résistent à la décomposition et s'accumulent dans le réseau alimentaire aquatique, atteignant souvent des concentrations élevés chez les animaux qui se nourrissent de poisson. Leurs effets toxiques sur les animaux de laboratoires, les animaux piscivores et les humains touchent notamment les systèmes reproducteur, immunitaire et nerveux. Malgré la réglementation de plusieurs POP depuis un quart de siècle, nombre de ces substances persistent dans l'environnement et continuent de poser un risque pour la santé des animaux sauvages qui se trouvent au sommet de la chaîne alimentaire. Des processus atmosphériques dispersent des POP au quatre coins du monde, ce qui s'ajoute à l'influence des sources régionales en Colombie-Britannique et dans les eaux adjacentes. Selon des données récentes, même des niveaux de contamination faibles à modérés perturbent des processus endocriniens chez des phoques communs de la Colombie-Britannique. L'observation selon laquelle les orques de la Colombie-Britannique comptent parmi les mammifères marins les plus contaminés au monde souligne le besoin de mieux comprendre les sources et le devenir des POP dans l'environnement ainsi que leurs effets sur la santé des organismes de niveau trophique élevé. L'utilisation de mammifères marins comme « sentinelles » fournit une mesure intégrative de la contamination de l'écosystème marin et de la chaîne alimentaire dont ils dépendent.

## 1. Introduction and Background

This paper summarises our current knowledge of Persistent Organic Pollutants (POPs) in Pacific marine mammals, with an emphasis on British Columbia. We focus on two aspects of the subject: trends (both spatial and temporal) in POP concentrations, and impacts in terms of sub-lethal effects. First, some definitions are in order.

We have taken “Persistent Organic Pollutants” (POPs) to describe compounds, or groups of compounds, which are composed of C, H and Cl, occasionally with O. They include the organochlorines, such as the *DDT-group of insecticides* (the parent compound *p,p'*-DDT and its metabolites *p,p'*-DDD and *p,p'*-DDE and occasionally the *o,p'*-analogues); the *polychlorinated biphenyls (PCB),* a group of 209 theoretically possible compounds, although fewer are found in environmental samples*;* the *polychlorinated dibenzo-p-dioxins (PCDD; n=75 compounds, including the most toxic “dioxin”, 2,3,7,8-TCDD) and dibenzofurans (PCDF; n=135 compounds)* which are usually analysed together and recorded together as PCDD/F. In addition, sporadic data exist for some other chlorinated pesticides such as *toxaphene, methoxychlor* and the *hexachlorocyclohexanes (HCH)* including *lindane*. Although polynuclear aromatic hydrocarbons (PAHs) are considered as POPs in some contexts, they do not bioaccumulate in marine mammals and are therefore not considered in this paper.

While approximately two dozen marine mammal species frequent British Columbia coastal waters, only a few have been studied. Most data in this paper refer to harbour seals (*Phoca vitulina*) and killer whales (*Orcinus orca*), both of which are resident in BC waters and have been studied recently. In this paper, we focus on marine mammals, but complementary research on other high trophic level biota is also available. Studies of fish-eating birds provide additional data for the interpretation of spatial and temporal trends, and adverse effects of POPs both in BC (Sanderson *et al.* 1994a;Sanderson *et al.* 1994b;Elliott *et al.* 1996a;Elliott *et al.* 1996b;Elliott *et al.* 2000;Elliott *et al.* 1996c) and in other regions, such as the Great Lakes (Gilbertson *et al.* 1991).

### *(a) Sources and environmental behaviour of POPs*

The DDT group of insecticides was used fairly extensively throughout Canada from the mid-1940's to the early 1970's, for various forms of insect control. PCBs were used in several industrial applications, typically as heat- and fire-resistant electric transformer oils and industrial lubricants, over about the same period. Both DDT and PCBs were effectively banned from manufacture and open use in North America and Europe during the early to mid-1970's. PCDD/F have been formed as by-products during various steps of pulp or paper manufacturing processes; until about 1989 when regulations and process changes reduced release to the environment. PCDD/Fs continue to be introduced to the BC environment as a consequence of incomplete, low-temperature combustion (incineration, vehicle exhaust), as well as lesser amounts from forest fires. Little is known about the extent to which these latter processes are relevant from a marine contamination perspective, although atmospheric inputs of many POPs have been documented in the Rocky Mountains (Blais *et al.* 1998). The use and improper disposal of PCBs and DDT continues in many developing nations, providing a source for ongoing global atmospheric distribution, although the large soil reservoir of PCBs emitted since the 1940s is probably the major factor controlling current environmental concentrations.

The environmental behaviour of POPs is largely controlled by three of their physical and chemical properties: *volatility, lipid solubility and chemical resistance*. First, the POPs are *volatile* --- they usually have measurable, but low, Vapour Pressures (VP) at normal environmental temperatures. A fraction of their distribution is therefore in vapour phase, leading to widespread dispersion by atmospheric transport. Prevailing winds in BC are from the west, suggesting that atmospheric sources of POPs to the BC marine environment may originate from trans-Pacific sources (e.g. Asia)(Wilkening *et al.* 2000). On the other hand, POPs released regionally (e.g., PCDD/F from coastal pulp mills, PCBs from Puget Sound sources) can either contribute via atmospheric processes to local deposition, or to more distant, interior regions to the east. Second, most POPs have *high lipid/water partition coefficients and low water solubility*; this means that they partition readily into lipid rich tissues (such as marine mammal blubber) or they adsorb readily to suspended particulate matter (SPM) and sediments. Finally, POPs are fairly *stable chemically* --- many POP compounds are highly resistant to enzymatic or abiotic breakdown, resulting in their extreme persistence in the food chain and in the environment.

*(b) Factors affecting POP distribution in marine mammals*

Although the physico-chemical properties of the POPs control much of their environmental behaviour, their dynamics and distribution in marine mammals are largely controlled by physiological and biochemical processes. POPs tend to be highly lipophilic, causing them to be distributed in fatty tissues throughout the body. The bulk of the POP burden is therefore associated with blubber in marine mammals, since this energy and insulating tissue represents the key lipid storage depot for marine mammals.

The main factors controlling lipid distribution in marine mammals are *feeding and reproduction*. The role of *feeding* is obvious, and the POP burdens accumulated by a marine mammal depend on the amount of food consumed, the efficiency of POP absorption, and the POP concentration in the food. The latter can vary with the trophic level of the diet, since POP concentrations generally increase with increasing trophic levels (Muir *et al.* 1988). We recently attributed the relatively high levels of PCBs in transient killer whales to their consumption of marine mammals, compared to the “less contaminated” fish-eating resident killer whales (Ross *et al.* 2000a).

Other than feeding, the main process affecting POP burdens in marine mammals is that of *lactation during reproduction*. Depending on the species, female marine mammals can mobilise up to 90% of their blubber to produce a lipid rich milk to sustain their offspring during nursing (Bowen *et al.* 1992). Studies suggest that the majority of the transfer from mother to offspring takes place via nursing and not transplacentally. Although the POPs associated with maternal blubber are not exported wholesale to the offspring, the mother loses (and the unsuspecting offspring gains) a significant fraction of maternal POP residues (Addison *et al.* 1977). Annual uptake (at least of the DDT-group) during feeding was shown to be approximately balanced by loss during lactation in Atlantic grey seals (Addison *et al.* 1977). Male marine mammals, on the other hand can rely only on slow metabolic degradation to "clear" POP burdens. Indeed, POP concentrations consistently increase in male marine mammals as they age (Addison *et al.* 1998;Addison *et al.* 1974;Ross *et al.* 2000a), and there is some data to suggest the same in non-breeding (i.e. post-reproductive) females (Ross *et al.* 2000a).

Concentrations of POPs recorded in marine mammals therefore vary with *sex, age and condition*. POP concentrations appear to vary inversely with blubber thickness, a measure of condition. The practical outcome of these considerations is that POP *concentrations* --- which are the data actually measured and recorded --- cannot be interpreted reliably unless these ancillary variables of *age, sex, reproductive status and condition* are also available (Addison 1989;Ross 2000).

## 2. Data sets examined

Until the early 1990's, there were very few studies of POP concentrations in BC marine mammals. Sporadic samples originating from individuals of variable quality and different species identified POPs in marine mammals from BC and Washington (Jarman *et al.* 1996). During the late 1960s and early 1970s, some studies were carried out by US scientists on Pribilof fur seals (Anas *et al.* 1970), California sea lions in Oregon (*Zalophus californianus californianus*: Buhler et al., 1975), and more recently for sea otters (Estes *et al.* 1997) and northern fur seals (Beckmen *et al.* 1999) in Alaska. A temporal trend for total PCBs and organochlorine pesticides was established for harbour seals in southern Puget Sound between 1972 and 1984 using earlier analytical techniques (Calambokidis *et al.* 1984;Calambokidis 1995;Calambokidis *et al.* 1991).

Since the early 1990's, samples of healthy, free-ranging and known–age harbour seals and killer whales have been taken from BC waters on a more systematic basis. Comparison with other studies is complicated to a degree by differences in analytical techniques, as well as the variable age, sex and condition of the animals being sampled in other studies. With that caveat, there follows a list of data sets that we have used for this paper.

*(a) Harbour seals*

(i) In 1991 and 1992, harbour seals were shot because they were preying on fish farms in the Strait of Georgia and in Quatsino Sd. Age, sex and season were recorded and samples of blubber were analysed at the IOS Regional Dioxin Laboratory (RDL) for 2,3,7,8-substituted PCDD/Fs and for a suite of mono-*ortho* and non-*ortho* substituted PCBs. Data have been summarised elsewhere (Addison *et al.* 1996;Addison *et al.* 2001).

(ii) During 1996 and 1997, weaned harbour seals were sampled from various sites in southern British Columbia (most from the Strait of Georgia). During the 1996 study, 24 seals were temporarily placed in pools at the Cultus Lake Salmon Research Laboratory for immunotoxicological assessment. As part of this study, blubber biopsy samples were taken for PCB, PCDD and PCDF analysis. In 1997, free-ranging seals of the same age class were live-captured for endocrine assessment, and a blubber biopsy sample taken for PCB, PCDD and PCDF analysis. These data are partially summarized elsewhere (Ross *et al.* 1998a;Simms *et al.* 2000a), with additional manuscripts currently in preparation.

(iii) During 1996-97, blubber biopsies were taken from 17 free-ranging, weaned harbour seals of the same age class as (ii) above as part of a collaborative project with the Washington Department of Fish and Wildlife (WDFW), the Puget Sound Water Quality Action Team (PSWQAT), Cascadia Research Collective (CRC), and the U.S. Environmental Protection

Agency (USEPA). Together with archived samples collected from “healthy dead” (i.e. normal blubber layer) harbour seal pups collected in 1984, 1990, and 1993, these were analyzed at IOS for full congener PCB, PCDD and PCDFs. This work has even greater temporal significance, since CRC had already analyzed the 1984 and 1990 samples, as well as samples dating back to 1972, using a simpler analytical technique. This work is summarized elsewhere (Calambokidis *et al.* 1999a;Ross *et al.* 1998b), with further submissions underway.

*(b) Killer Whales*
(i) Between 1993-1996, 47 skin and blubber samples were taken by biopsy dart from three populations (northern residents, southern residents and transients), all of known age, sex and lineage. Samples were analysed for full congener PCB, PCDD and PCDFs; data have been analysed and published (Ross *et al.* 2000a).

(ii) Six stranded (i.e. deceased) killer whales of varying ages were collected during the late 1980's and were analysed for PCDD/F, a range of POP pesticides including the DDT-group, and for selected PCB congeners (Jarman *et al.* 1996).

## 3. Spatial variation in POP residues

*(a) Harbour Seals: Strait of Georgia vs. Quatsino Sound*
Harbour seals in the Strait of Georgia are strongly philopatric and generally remain within an area of 50 $km^2$ (Cottrell 1998). An examination of the 1992 sampling of Strait of Georgia and Quatsino Sd. harbour seals shows some clear differences in PCDD/F concentrations. Although it was not possible to control for biological factors during the sampling, male animals from the two sites (n=5 and 7, respectively) did not differ significantly in age (8.60 ± 5.08 and 5.57 ± 3.31 years respectively, mean ± s.d.). Blubber thickness did differ significantly (t-test), with the Strait of Georgia samples being about half as fat as the Quatsino Sd. samples (16.2 ± 6.76 and 29.4 ± 13.8 mm respectively). However, total PCDD concentrations were about 20-fold higher in the Strait of Georgia samples than in Quatsino Sd. (Fig. 1), and males had consistently higher concentrations than females. Similar spatial differences were seen in PCDF concentrations, with the Strait of Georgia samples having about 5-fold higher PCDF concentrations than in Quatsino Sd. (Fig. 2). Allowing for the differences in condition between the two samples, it seems that Strait of Georgia harbour seals (in 1991-92) had about 10-fold higher PCDD/F burdens than those in Quatsino Sd.

Closer examination of the distribution of PCDD/F congeners showed that much of the difference between the two sites could be attributed to two compounds, 1,2,3,6,7,8-HxCDD, and 2,3,7,8-TCDF. 1,2,3,6,7,8-HxCDD represented a significantly higher percentage of the total PCDD in Strait of Georgia than Quatsino Sd. samples (87.2 ± 15.2 % vs. 58.9 ± 4.7 %, respectively). Similarly, 2,3,7,8-TCDF represented a higher percentage of PCDF in Strait of Georgia samples than in Quatsino Sd. (89.0 ± 7.4 % vs. 73.6 ± 16.5 %, respectively).

These spatial differences are almost certainly due to the influence of pulp and paper mill processing discharges, which contained appreciable amounts of PCDD/F until the early 1990's. The Strait of Georgia received direct inputs of such discharges from six mills which produced bleached kraft mill effluent (BKME) plus, indirectly, the discharges via the Fraser River of mills at Prince George, Quesnel and Kamloops. Many of the mills used a feedstock consisting of wood chips preserved with pentachlorophenol, which during processing formed by-products including 1,2,3,6,7,8-HxCDD (Macdonald *et al.* 1992). Other processes yielded 2,3,7,8-TCDD as a by-product, and the enrichment of these two compounds in the Strait of Georgia samples provides a convincing "fingerprint" of BKME exposure. In contrast, Quatsino Sd. samples were exposed to effluent from a sulphite mill at Port Alice which has used only mild chlorine bleaching and a feedstock free of PCDD/F precursors (Yunker *et al.* 1996).

Concentrations of total non-*ortho*-PCB (NO-PCB) in Strait of Georgia samples were about twice those in samples from Quatsino Sd. and the difference was accounted for mainly by the pentachloro- isomers #126 and 127. PCB 77 represented about 60% of the tetrachloro- NO-PCB, and PCB 127 about 80% of the pentachloro-NO-PCB. Since the former animals were thinner, NO-PCB burdens were probably fairly similar at both sites. However, PCB 77 appeared to represent a smaller proportion of total NO-PCB in Strait of Georgia than in Quatsino Sd. samples, for unknown reasons.

Total mono-*ortho*-PCB (MO-PCB) concentrations were about 3 - 5 fold higher in Strait of Georgia samples than in Quatsino Sd., and most of this difference was due to the pentachloro substituted isomers, of which PCB 118 represented close to 75% and PCB 105 a further 20%. (Note that concentrations of these compounds were 1000-fold higher than those of the PCDD/F or NO-PCB.) There was no obvious enrichment of any of the MO-PCBs in any group of samples. However, after allowing for differences in mean blubber thickness (which would explain a two-fold difference in concentrations) site differences were fairly small and the simplest interpretation of the PCB data would be that the slightly higher burdens in the Strait of Georgia samples reflect PCB inputs over a relatively long period associated with general industrial and urban activity.

*(b) Harbour seals: BC vs. northern Europe*

PCDD/F have been analysed in other harbour seals. Five harbour seals from an unspecified site in the North Sea (sampled in response to the 1988 *Morbillivirus* epizootic) had mean total PCDD plus PCDF concentrations in the 10-30 pg/g blubber lipid range (Bergek *et al.* 1992). In the Baltic Sea, five major 2,3,7,8-substituted PCDD and PCDF accounted for 21-46 pg/g blubber lipid in pools of several individuals of varying age collected between 1979 and 1990 (Bergek *et al.* 1992); this is slightly higher than the range of values found in Quatsino Sd. samples, but well below those in the Strait of Georgia. Harbour seals from the Swedish west coast also had total PCDD and PCDF concentrations around 20 pg/g blubber lipid or less, but 1,2,3,6,7,8-HxCDD was apparently not detected in these samples (Bignert *et al.* 1989). Thus, harbour seal samples from Quatsino Sd. appeared to be contaminated with PCDD and PCDF at similar concentrations to those reported from northern Europe, while Strait of Georgia harbour seals contained PCDD/F concentrations well above those reported elsewhere.

PCB levels appear to be much lower in BC harbour seals than in young animals sampled in northern Europe, including the heavily contaminated Baltic Sea (Ross *et al.* 1996a), although variable age and condition of seals sampled in European studies make a direct comparison difficult.

*(c) Harbour seals: BC vs. Puget Sd.*
Based on blubber biopsy samples taken from healthy, same-age, free-ranging harbour seals from both the Strait of Georgia and southern Puget Sound in 1996-97, clear differences in contaminant levels are evident. While inter-site total PCB levels differed little within the Georgia Basin, harbour seals from southern Puget Sound were approximately five times more contaminated than their Strait of Georgia counterparts (Simms *et al.* 2000a;Ross *et al.* 1998b). However, total PCDD and PCDF concentrations were higher in the Strait of Georgia seals, likely reflecting proximity to numerous pulp mills.

Continuing collaboration with WDFW, PSWQAT, CRC and USEPA aims to further substantiate spatial and temporal differences in contaminant levels and sources in the upper trophic levels of the marine food chain in British Columbia and Washington.

*(d) Harbour seals: BC vs. eastern Canada*
Concentrations of all PCB congeners analysed (both sites, both sexes) were around 0.5 µg/g wet wt., but it is difficult to compare these data with other values because of the differences in methods of PCB analysis used, and in the suite of congeners selected. For what the comparison is worth, however, a suite of blubber samples from harbour seals taken at Sable Is. in the late 1980's contained PCB congeners in the range 0.5 - 1.5 µg/g (R.F. Addison and W.T. Stobo, unpublished data). It should be noted that the suite of congeners analysed in the Sable Is. samples was larger than that analysed in the Strait of Georgia and Quatsino Sd. Samples, and that age differences may also affect this comparison.

In conclusion:
- PCB concentrations in blubber of BC harbour seals sampled in 1991-92 seem roughly similar to those in harbour seals from Atlantic Canada, but appreciably lower than those in samples from southern Puget Sd.
- PCDD/F concentrations in seal samples from a "reference" site at Quatsino Sd. were similar to concentrations reported from northern European waters at about the same time.
- Concentrations of PCDD/F from Strait of Georgia harbour seals were about 10-fold higher than at the Quatsino Sd. site, with the pattern of congeners representing a strong pulp mill effluent “fingerprint".

*(e) Killer whales: inter-population differences*
BC killer whales fall into three identifiable populations: *northern residents* (Georgia and Johnstone Straits, western Vancouver Is., central coast N of about 50ºN to the Alaska border during the summer months); *southern residents* (Georgia Strait, Juan de Fuca Strait, western Vancouver Is. and Puget Sound S of about 50ºN), and *transients* (elusive, but frequent coastal waters of Washington, BC and Alaska). The two resident groups are mainly fish eaters (strong preference for salmon); the transients are marine mammal eaters (Ford *et al.* 1998). Because of the wide geographic range of these animals and their prey, they cannot be considered good indicators of *local* contamination. However, their very high trophic level, coupled with their long lives, make them very good *integrators* of food chain contamination in British Columbia and the northeast Pacific Ocean. It should be noted that BC’s resident killer whales represent one of the few cetacean population in the world with fully identifiable, known-age individuals. The existence of an invaluable extensive photo-identification catalogue has formed the basis for the interpretation of contaminant

concentrations in these free-ranging mammals, as well as for numerous other studies (behaviour, feeding ecology, distribution, and genetics) (Ford *et al.* 1994;Ford *et al.* 1999).

Biopsy dart samples of blubber taken from 26 northern residents, 15 transients and 6 southern residents between 1993-96 (sampled by Graeme Ellis, Pacific Biological Station, and Lance Barrett-Lennard, PhD candidate at the University of British Columbia) were analysed for PCDD/F and 156 PCB congeners (136 were detected in all whales sampled) (Ross *et al.* 2000a). While PCDD/F concentrations did not differ among the groups, PCB concentrations were significantly higher in the transient males than in southern resident males; northern resident males had intermediate concentrations (Fig. 3). It has previously been established that PCBs are highly persistent in marine mammals, whereas PCDDs and PCDFs are more readily metabolized (this is not the case in humans) (Boon *et al.* 1997).

The PCB concentrations found in all three killer whale populations are very high. The southern resident and transient killer whales are three to five times more contaminated with this class of industrial chemical than the endangered St Lawrence Beluga whale (Muir *et al.* 1996). This places BC's killer whales among the most contaminated marine mammal populations in the world (based on a more limited analytical technique, the Mediterranean striped dolphin would be considered on a par with the transients). While the high PCB levels in transients partly reflects their high trophic level, it would appear that a slightly divergent prey selection could explain the contamination of southern residents relative to northern residents. This could reflect the occasional consumption of more contaminated locally inhabiting fishes in e.g. Puget Sound area, or the consumption of larger amounts of higher trophic level species. Further research is underway to characterize the origin of these PCBs for killer whales.

In conclusion:

- PCDD/F concentrations show no clear variation among the three groups of killer whales (northern residents, southern residents and transients) sampled in BC coastal waters, possibly reflecting a lack of bioaccumulation in these high trophic level animals and metabolic elimination.
- PCB concentrations were higher in transient males than in resident males, probably reflecting dietary differences between the groups.
- PCB levels in the three killer whale populations are very high, being matched only by the Mediterranean striped dolphin, and may be due to a combination of regional inputs and international (atmospheric) sources of POPs into the northeast Pacific food chain.

## 4. Temporal trends in POP concentrations

No historical data exist from British Columbia to make inferences about temporal trends in POPs in high trophic level marine mammals. From studies carried out on populations from other areas, such as the western Arctic ringed seals (Addison *et al.* 1986;Addison *et al.* 1998) and Sable Is. grey seals (Addison *et al.* 1984;Addison *et al.* 2000), we know that seal populations have responded slowly --- over decades --- to changes in the environmental input of the DDT-group of insecticides and the PCBs. In addition, while clear reductions in the levels of PCBs were observed in biota in North America and Europe between the early 1970s and the mid 1980s, virtually no changes have been observed over the past 15 years (Calambokidis *et al.* 1999a). This suggest that while regulations proved initially effective in reducing the release of PCBs into the environment, these chemicals continue to circulate as a result of i) continued leakage from old storage sites; ii) cycling within environmental compartments; and iii) atmospheric inputs from distant sources (e.g. Asia, USA, Europe).

*(a) PCDD/F trends in Strait of Georgia harbour seals*

Since PCDD/F discharges to the Strait of Georgia have declined dramatically since about 1990, the question arises as to whether these changes are reflected in residue concentrations in marine mammals. Because seals have been shown to be able to metabolically remove dioxin-like compounds (De Swart *et al.* 1995;Boon *et al.* 1997), levels of PCDDs and PCDFs might be expected to decrease more rapidly than the more persistent PCBs once the source is removed.

PCDD/F concentrations in harbour seal pups collected from the southern Strait of Georgia in 1992 (Addison et al., 1996) were compared with those in post-weaning pups biopsy sampled from the same area in 1996. Two "marker" compounds, 1,2,3,6,7,8-HxCDD and 2,3,7,8-TCDF which are representative of pulp mill effluents (Yunker and Cretney 1996) had declined significantly ($P < 0.01$ by *t*-test) over the interval (Fig. 4; Addison and Ross, unpublished data).

Although the marine mammal temporal trend data are sparse, they appear consistent with sediment core data (Macdonald *et al.* 1998;Bright *et al.* 1999) and data from Great blue herons and other seabirds (Elliott *et al.* 1996b;Elliott *et al.* 1996d;Sanderson *et al.* 1994b). The general picture is one of increasing input of POPs into the SE BC ecosystem from the 1940s onwards as a result of industrial activity or deliberate (pesticide) spraying. After the early 1970s, local sources of DDT and PCB were controlled (although atmospheric inputs from distant sources have continued) and a slow decline in POP concentrations in various environmental compartments began. PCDD/F inputs were controlled after 1990, and this seems to be reflected in declines in the concentrations of these chemicals in more recent samples (taken during the 1990s).

*(b) PCB trends in southern Puget Sound harbour seals*

Consistent with observations in biota elsewhere in the industrialized world, levels of PCBs and DDT in harbour seals from southern Puget Sound declined dramatically between the early 1970s and the mid 1980s, with no change being observed over the past 15 years (Calambokidis *et al.* 1999b). This would suggest that these chemicals continue to be introduced into the regional environment, and are not degrading as rapidly as had once been hoped.

In conclusion:

- Concentrations of 1,2,3,6,7,8-HxCDD and 2,3,7,8-TCDD ("marker" congeners of pulp mill effluent discharges) declined significantly in Strait of Georgia harbour seal pups between 1992 and 1996, reflecting reductions in PCDD/F discharges from coastal pulp mills after 1990.
- While PCB levels in environmental samples dropped following the implementation of regulations 25 years ago in Canada and the U.S., there has been little change in concentrations in Puget Sound seals since 1984, suggesting the continued presence of this toxic chemical in our coastal food chain.

## 5. Contaminant-related toxicity and risk of toxic effects in marine mammals

While the presence (i.e. *detectability*) of anthropogenic contaminants in the natural environment is of concern to society, their presence at levels which can cause toxic injury (i.e. *toxicity*) represents a more relevant regulatory perspective. It was therefore a surprise to discover PCBs and DDTs in samples collected from remote regions of the world in the mid 1960s (Jensen 1966), but of greater concern to managers were the catastrophic reproductive failures of many fish-eating bird species throughout North America and Europe in the late 1960s and early 1970s as a consequence of DDT-associated eggshell thinning (Hickey *et al.* 1968). The extirpation of Bald eagles, terns, cormorants, Peregrine falcons and other species from wide tracts in Canada and the U.S. provided tangible evidence of population-level effects in high trophic level wildlife.

As with studies of humans, demonstrating a mechanistic or "cause and effect" relationship between POPs and adverse health effects in marine mammals has proven challenging. Logistical, ethical and legal constraints have made it difficult to obtain conclusive data on the toxicity of specific chemicals. The conserved nature of many physiological processes among mammals (and other vertebrates) enables extrapolation between species, forming the basis for much work in toxicology (e.g. the use of laboratory rodents in assessing risk to human health from chemicals or drugs). A "weight of evidence" based on the accumulated results of laboratory animal studies, captive marine mammal studies, and studies of free-ranging marine mammals, provide evidence that ambient environmental levels of POPs present a continued risk to several populations of different marine mammal species (Ross 2000;Ross *et al.* 2000b). The "weight of evidence" approach maximizes ecological relevance while strengthening our understanding of mechanism of toxic action and identifying putative chemical agents responsible for effects (Fig. 5). Abundant epidemiological evidence of contaminant-related effects exists for marine mammal populations inhabiting the Baltic Sea, the Mediterranean Sea, the Japanese coast, the St Lawrence estuary, the Gulf of Mexico, coastal California and Puget Sound (Helle *et al.* 1976;De Guise *et al.* 1995;Aguilar *et al.* 1994;Delong *et al.* 1973;Lahvis *et al.* 1995;Subramanian *et al.* 1987;Reijnders 1980), although few studies have adequately addressed high trophic level wildlife in the developing world.

Two captive feeding studies of harbour seals in the Netherlands provides some of the strongest direct evidence to date that POPs in the environment are adversely affecting free-ranging marine mammals. In the first of these, seals were fed either a diet of fish from the contaminated Rhine estuary, or fish from the cleaner eastern Wadden Sea (Reijnders 1986). Female reproduction was adversely affected, with fewer pups produced and an absent oestradiol peak during the reproductive season. Circulating levels of both thyroid hormone (TT4) and vitamin A were reduced, with laboratory animal studies attributing this to the disruption of the common transport complex for these two endocrine factors (Brouwer *et al.* 1989). A second captive study was designed to assess the possible role of POPs in the virus-associated mass mortality of 20,000 harbour and grey seals in northern Europe in 1988. Harbour seals were fed either herring from the contaminated Baltic Sea or the relatively uncontaminated Atlantic Ocean. During a 30-month study, blood samples were regularly taken for immunological assessment. Seals in the Baltic group exhibited diminished T-cell function in vitro and in vivo, as well as reduced natural killer function (De Swart *et al.* 1994;Ross *et al.* 1995;Ross *et al.* 1996b). Seals in the Baltic group accumulated 17 mg/kg PCBs (209 ng/kg TEQ for total dioxin-like PCBs, PCDD/Fs) by the end of the feeding study, a figure since used as a "threshold" for immunotoxicity and endocrine disruption (Ross *et al.* 1995). The evidence for a specific effect of POPs was strengthened by two parallel studies using laboratory rodents exposed to the complex mixture of POPs found in the two herring batches; the pattern of immunological effects was similar in rats and seals (Ross *et al.* 1996c;Ross *et al.* 1997). The authors concluded that contaminants likely contributed to the severity of the virus outbreak by diminishing host resistance, increasing infection and transmission rates, and contributing to more serious disease (Ross *et al.* 1996a).

*(a) British Columbia and Puget Sound harbour seals*

We have recently been studying the relationship between environmental contaminants and biological endpoints in free-ranging harbour seals in British Columbia and Washington. Research (1996 to present) has focussed on understanding the POP concentrations from a spatial and temporal perspective, but this has been combined with efforts to documents effects and sources of POPs. We have documented a contaminant-related disruption of vitamin A (retinol) in harbour seal pups sampled in Washington and British Columbia (Fig. 6), suggesting that exposure to even low to moderate (2 to 17 mg/kg total PCBs) contaminant concentrations are affecting marine mammals in this region (Simms *et al.* 2000a). This observation may have population-level consequences because vitamin A is essential to normal growth and development, immune function and reproduction. Understanding and describing the confounding factors of age, nutrition and condition were key to discerning the effect of POPs on vitamin A physiology in this study, and provided much needed baseline information (Simms *et al.* 2000b).

Harbour seal pups in southern Puget Sound have an average total PCB concentration which is approximately equal to the previously established threshold for immunotoxicity and endocrine disruption from captive feeding studies (Ross *et al.* 1998b). Since adult seals (males in particular) will have higher burdens of these contaminants, the risk of adverse effects remains high in Puget Sound populations of seals, in particular.

*(b) Killer whales*

We do not have direct evidence that current POP concentrations are having an adverse effect on free-ranging killer whales in British Columbia. However, most individuals easily exceed the previously established toxic threshold established for harbour seals. These levels likely represent a health risk to killer whales; COSEWIC cited three stressors upon listing resident killer whales as “threatened” and transients as “vulnerable”: i) diminishing salmon returns (preferred prey for residents); ii) heavy vessel traffic (stress; interference with foraging); and iii) high contaminant levels (risk for reproductive impairment, immunotoxicity, disease, development). We are currently developing techniques to assess the effects of POPs on killer whale health, with some of these being validated in a harbour seal model.

## 6. Conclusions

- From a geographic perspective, it would appear that Puget Sound harbour seals are at the greatest risk for POP-related effects, reflecting their proximity to PCB sources and the contamination of the local food chain.
- Even though southern BC harbour seals are less contaminated with PCBs than their Puget Sound counterparts, there now exists evidence of a contaminant–related disruption of endocrine processes (vitamin A) in BC seals.

- While interspecies differences in sensitivity exist, a "weight of evidence" suggests that BC's killer whales may be at risk from the toxic effects of POPs.
- While work is needed to further characterize the many POPs in high trophic level animals in BC, evidence from other parts of the world, and the pattern of PCBs, PCDDs and PCDFs in marine mammals in this region, suggest that the PCBs present the greatest toxic risk to BC's marine mammals.

**Recommendations:**

- ➢ Research should be carried out to further characterize the concentrations, patterns and types of POPs in BC marine mammals in a spatial and temporal context (e.g. samples collected every five years from selected sites; continued collaboration with Washington State).
- ➢ Research should be carried out which addresses the issue of food chain contamination in order to document bioaccumulation (trophic level) and source (point, regional, and international sources; atmospheric transport and deposition; local vs offshore sources of POPs) issues.
- ➢ Research should continue to assess the risk of adverse effects on marine mammals by studying relevant biological endpoints, while minimizing the influence of confounding factors (e.g. age, sex and condition).
- ➢ Results of this research should be linked to past regulatory implementations in BC and Canada, but should also be related to current practices where regulations may be lacking (new chemicals, pharmaceuticals, surfactants).
- ➢ Given the evidence of long-range transport of atmospheric POPs, Canada should use such information at the current round of negotiations for phasing out the use of 12 priority POPs around the world (UN Convention on POPs).

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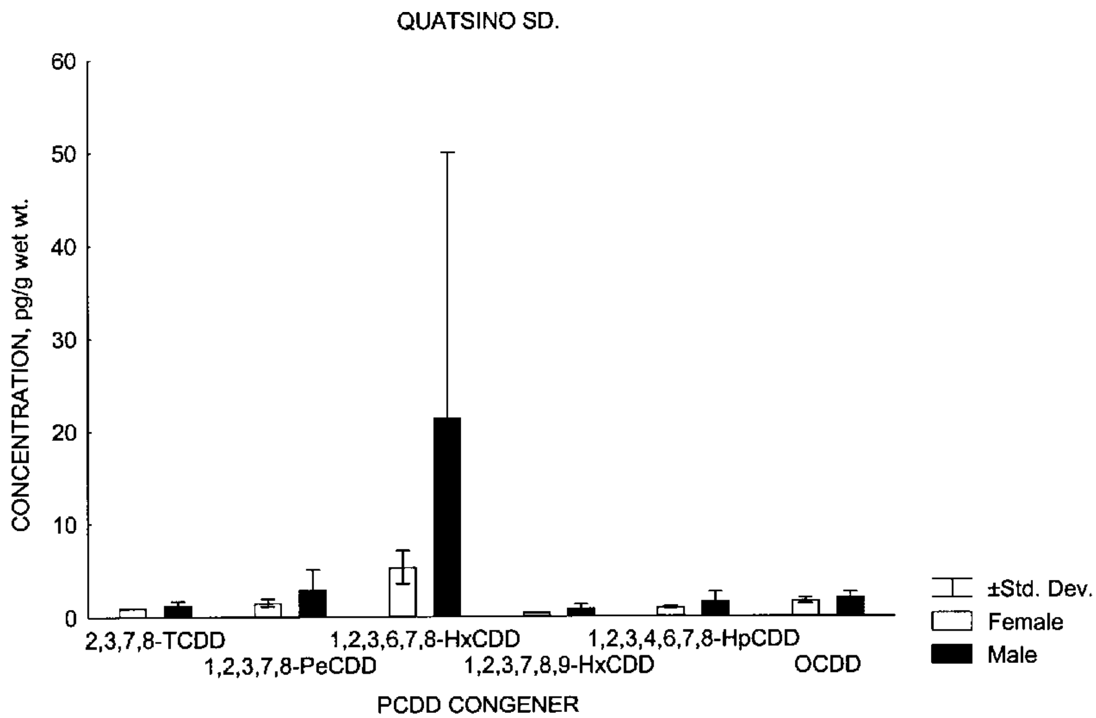
**Figure 1:** Higher dioxin concentrations in harbour seals sampled in the Strait of Georgia compared to Quatsino Sound reflect the historical inputs of pulp and paper effluents prior to regulations in 1989.



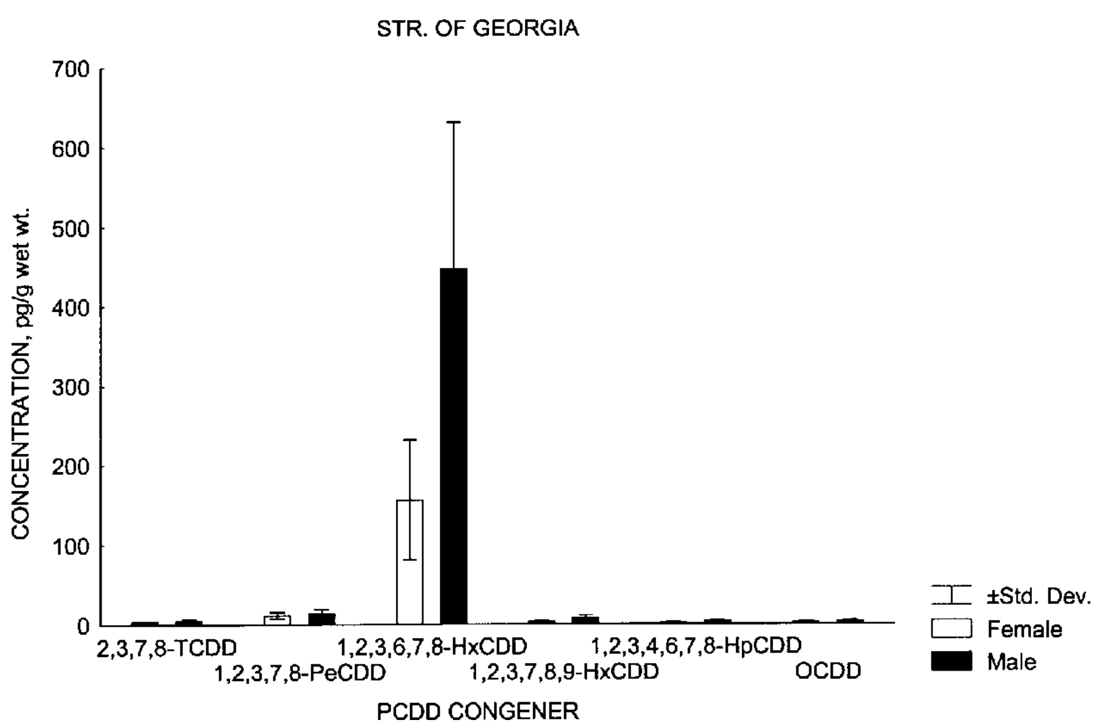
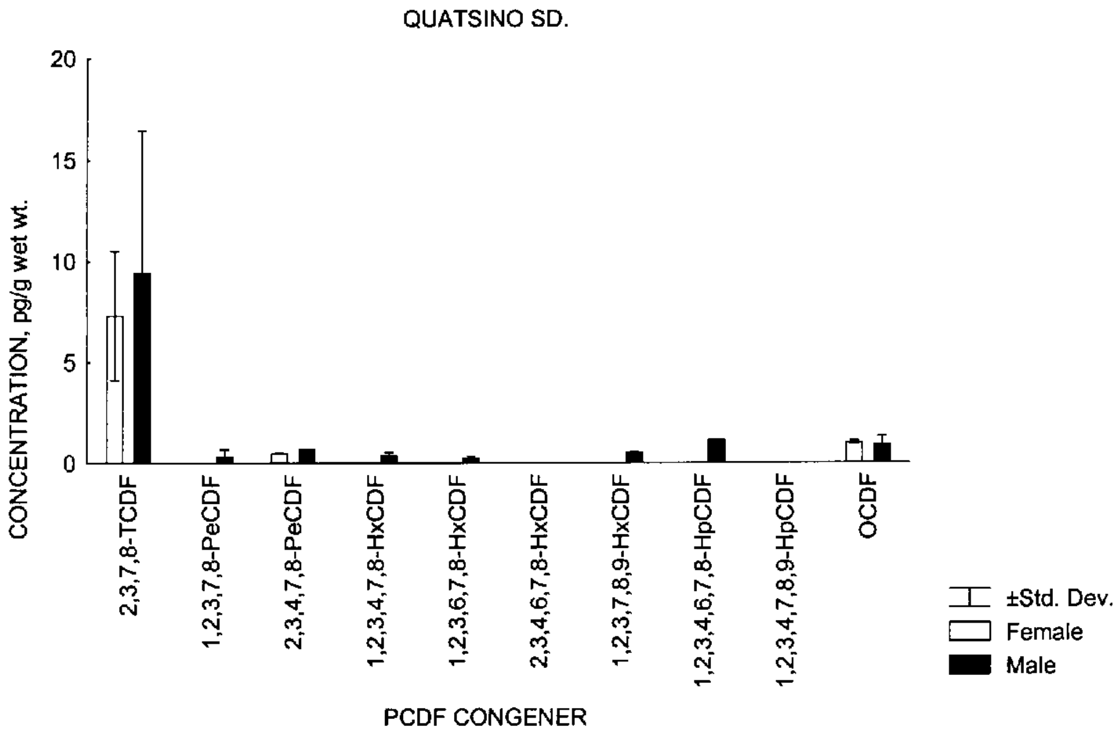



**Figure 2:** Higher furan concentrations in harbour seals sampled in the Strait of Georgia compared to Quatsino Sound reflect the historical inputs of pulp and paper effluents prior to regulations in 1989.



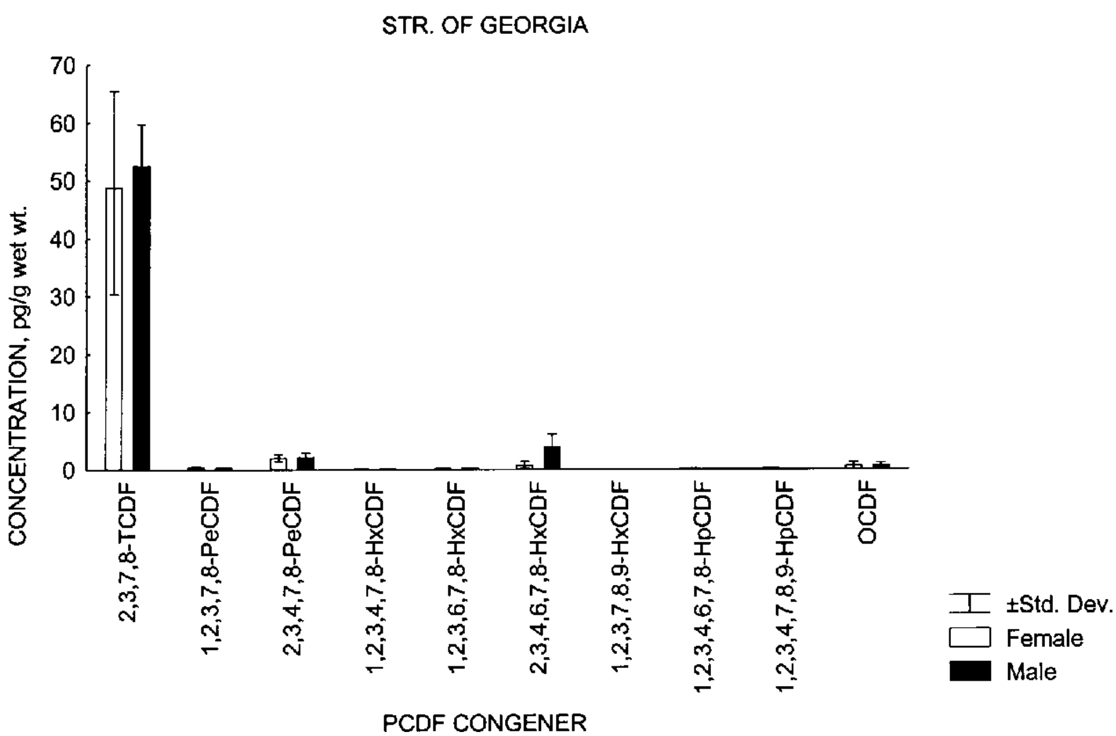



**Figure 3:** Total PCB concentrations were highest in the marine mammal-eating transient killer whales, but elevated levels are also of concern in the southern residents. Total PCDD and PCDF concentrations did not differ among populations or between males and females, suggesting that these compounds were not as persistent in killer whales as the PCBs (from Ross et al., 2000).

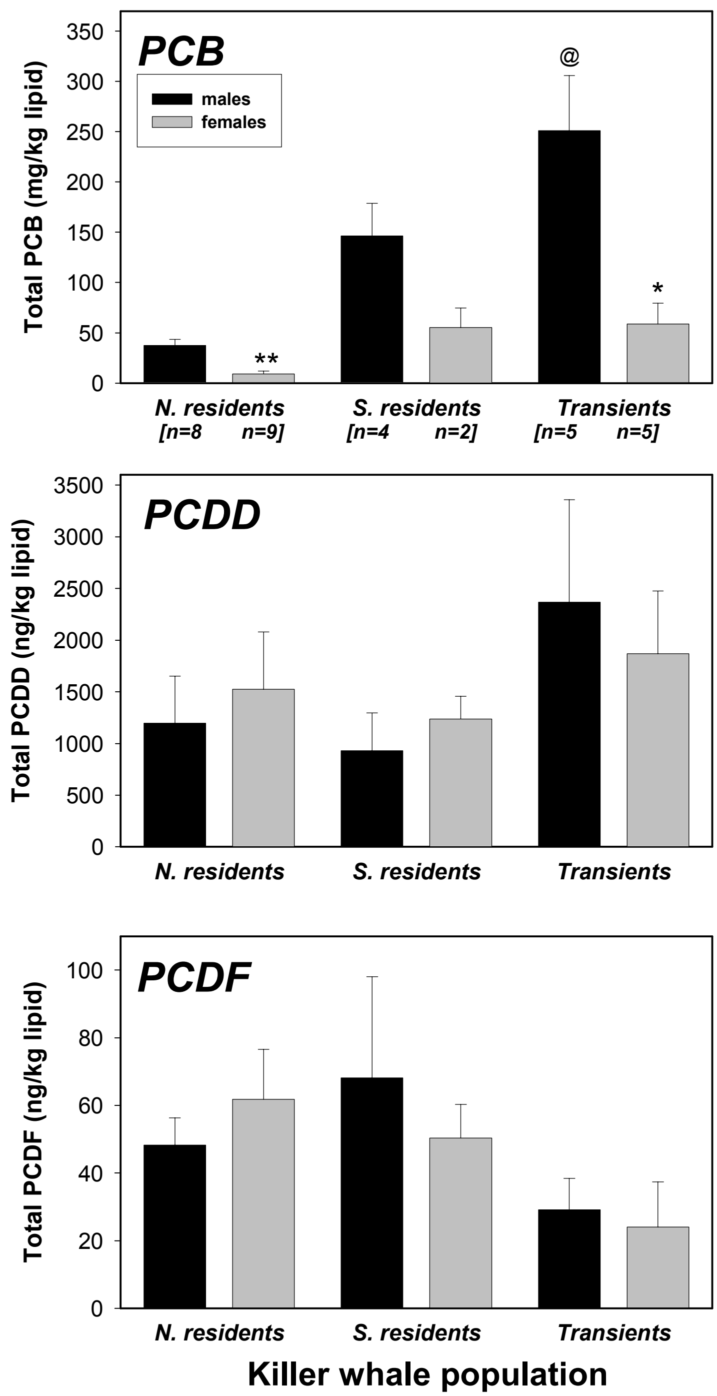
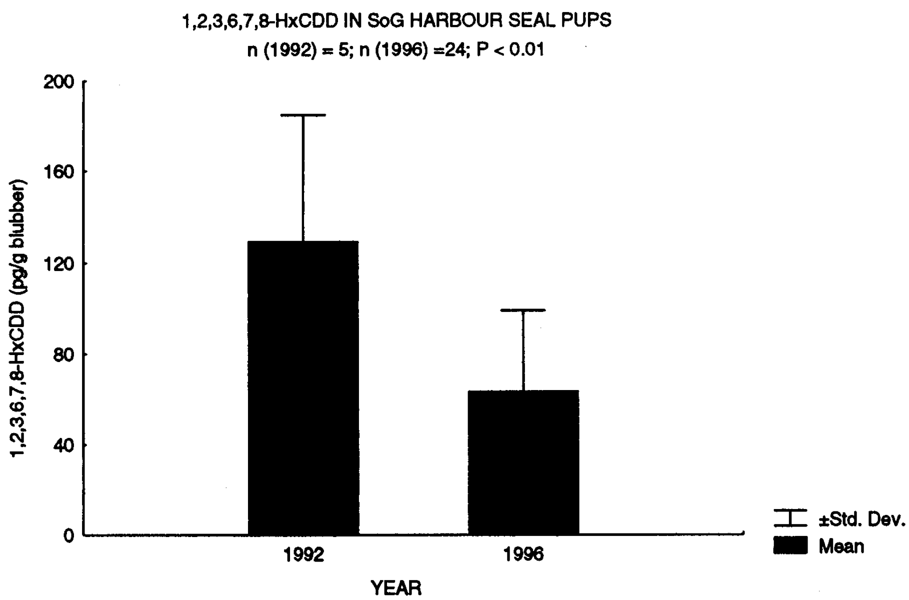


**Figure 4:** Declines in the concentrations of two pulp mill effluent "fingerprint" dioxin and furan congeners in Strait of Georgia harbour seals between 1992 and 1996 reflect the results of regulations enacted in 1989.



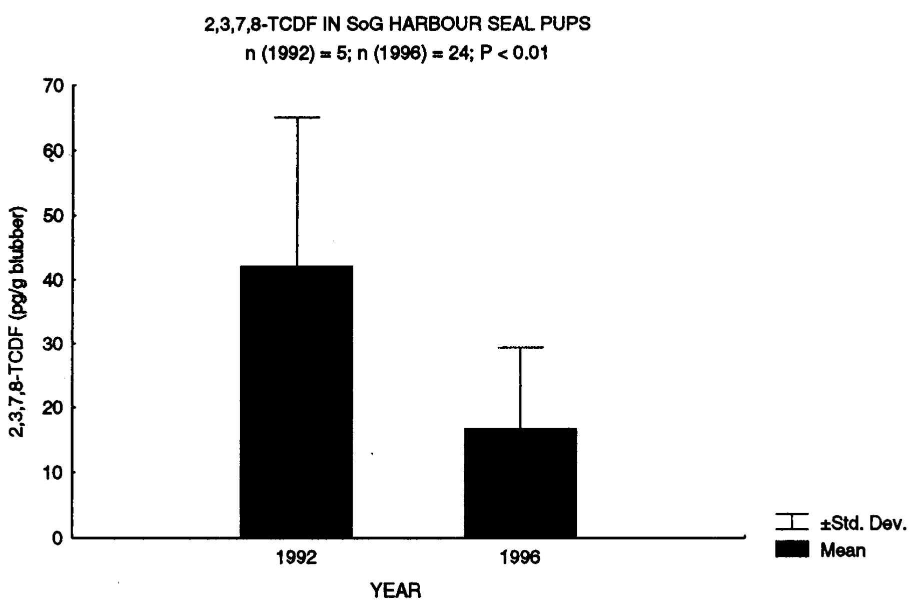



**Figure 5:** As with human studies of human health, legal, ethical and logistical constraints exist for studies of marine mammals. In order to maximize the ecological relevance and mechanistic understanding of marine mammal research, a "weight of evidence" approach represents a useful means of assessing impacts of POPs (from Ross, 2000).

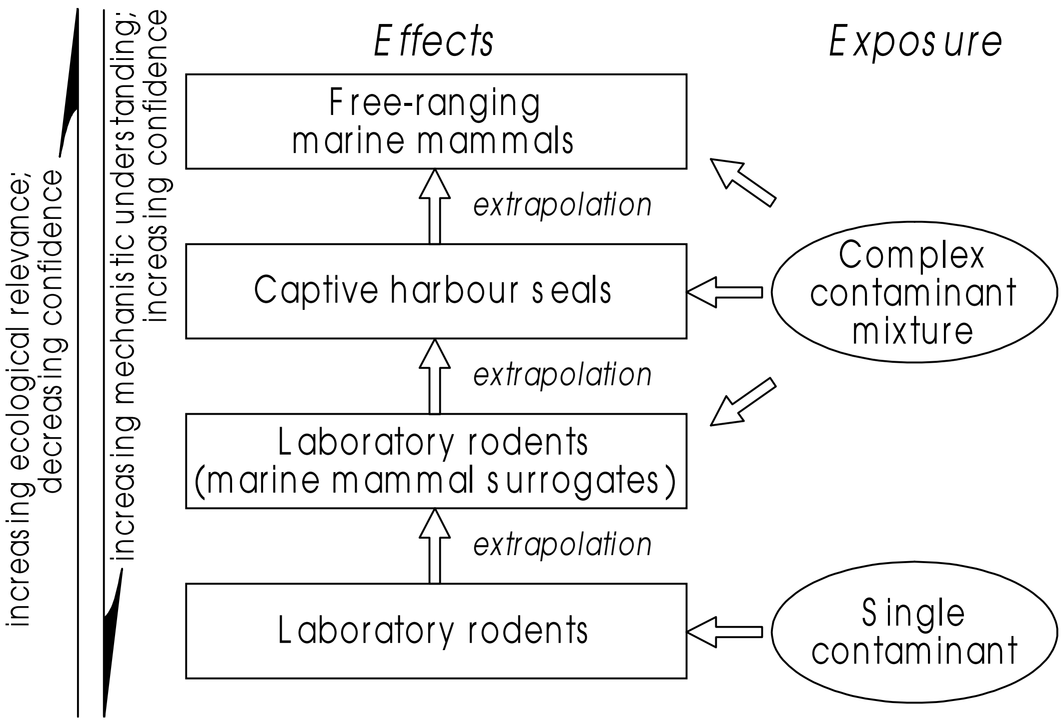


**Figure 6:** A contaminant-related disruption of retinol physiology was observed in both Puget Sound and Strait of Georgia harbour seal pups, suggesting that even low levels of exposure can have adverse health effects. Depleting liver stores were suspected to have resulted in this positive correlation between contaminant exposure (total TEQ and total PCB), something that has been documented in laboratory animals and fish-eating birds (from Simms et al., 2000).