Chlorinated Dioxin Trends between 1987 and 1993 for Samples of Crab Hepatopancreas from Pulp and Paper Mill and Harbour Sites in British Columbia

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ABSTRACT

Yunker, M. B., and W. J. Cretney. 1995. Chlorinated dioxin trends between 1987 and 1993 for samples of crab hepatopancreas from pulp and paper mill and harbour sites in British Columbia. Can. Tech. Rep. Fish. Aquat. Sci. 2082: xiii + 138 p.

Principal components analysis (PCA) has been applied to a large data set of the concentrations of chlorinated dibenzo-*p*-dioxins and dibenzofurans in samples of crab hepatopancreas from British Columbia (B.C.), Canada. These samples have been collected since 1987 from locations in the vicinity of B.C. coastal mills and the principal B.C. harbours. PCA distinguishes harbour samples from mill samples, reveals differences between individual mill sites and reliably classifies the dioxin and furan congener patterns according to chlorine bleaching, digested polychlorinated phenol and pentachlorophenol wood preservative sources. PCA is also used to assign sources to individual crab samples. In most cases changes in mill process chemistry since 1987 have produced dramatic reductions in both the proportion of toxic 2,3,7,8-chlorinated congeners and the overall chlorinated dibenzo-*p*-dioxin and dibenzofuran concentrations. The mill related tetrachlorodibenzofurans have been removed faster than the hexachlorodibenzo-*p*-dioxins, with the result that composition profiles and PCA projections for the mill sites have become more similar over time. Crab hepatopancreas samples from B.C.'s harbours have lower proportions of the toxic 2,3,7,8-chlorinated congeners, but have shown less change over time.

Keywords: chlorinated dibenzo-*p*-dioxins, chlorinated dibenzofurans, crab hepatopancreas, principal components analysis

RÉSUMÉ

Yunker, M. B., and W. J. Cretney. 1995. Tendances de la distribution des dioxines chlorées entre 1987 et 1993 dans des échantillons d'hépatopancréas de crabes provenant de sites d'usines de pâtes et papier et de ports de Colombie-Britannique. Can. Tech. Rep. Fish. Aquat. Sci. 2082: xiii + 138 p.

L'analyse en composantes principales (ACP) a été appliquée à un vaste ensemble de données sur les concentrations de dibenzo-*p*-dioxines et de dibenzofurannes chlorées présentes dans les échantillons d'hépatopancréas de crabes prélevés en Colombie-Britannique (C.-B.), Canada. Ces échantillons ont été recueillis depuis 1987 à proximité des usines de pâtes et papier de la côte et des ports importants. L'analyse permet de distinguer les échantillons recueillis à proximité de ports de ceux recueillis à proximité des usines. Elle révèle également les différences entre les sites de collecte à proximité des usines et classe de façon assez sûre les distributions des congénères dioxines et furannes en fonction du blanchiment au chlore, des phénols polychlorés digérés et des sources d'agents de protection du bois à base de pentachlorophénol. L'ACP peut également servir à déterminer les sources des différents échantillons de crabes. Dans la majorité des cas, les modifications apportées au procédé chimique des usines depuis 1987 ont induit des réductions drastiques à la fois dans la proportion des congénères toxiques, 2,3,7,8-dibenzo-*p*-dioxines, et de l'ensemble des congénères ainsi que des concentrations des dibenzofurannes. Les tétrachloro-dibenzofurannes provenant de ces usines ont diminué à un rythme plus rapide que celui des hexachloro-dibenzo-*p*-dioxines, faisant en sorte que les profils de composition et les prévisions de l'ACP sont graduellement devenus plus similaires à proximité des usines. Les échantillons d'hépatopancréas provenant des ports de C.B. montraient des proportions plus faibles de congénères tétrachlorées (2,3,7,8) mais un changement temporel plus lent que les autres congénères.

Mots-clés: dibenzo-*p*-dioxines chlorées, dibenzofurannes chlorées, hépatopancréas de crabe, analyse en composantes principales

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ACKNOWLEDGMENTS

Collection and data analysis for the pulp mill sites which constitute most of the samples in this study was performed by Hatfield Consultants Ltd., Beak Consultants Ltd., G3 Consulting Ltd. and Norecol Ltd. with chemical analyses done by Axys Analytical Ltd., Envirotest Laboratories Ltd. and Wellington Laboratories Ltd. All of this work was funded directly by the operators of the B.C. mills. Samples from remote areas and the B.C. harbours were collected by the Departments of Fisheries and Oceans and Environment and were primarily analysed by government laboratories.

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1. INTRODUCTION

Chlorinated contaminants produced and discharged by pulp mills were documented in British Columbia, Canada (B.C.) coastal waters more than a decade ago (e.g., Voss and Yunker 1983). More recently the presence of chlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in ecologically significant concentrations in sediments, shellfish and groundfish was described by Norstrom *et al.* (1988), both in the vicinity of B.C. mills and in the principal B.C. harbours. As one result, harvesting of prawn, shrimp and crab was restricted near pulp mills in late 1988 — more extensive fisheries closures have followed as the extent of contamination has been more fully delineated.

The major processes that produced PCDD/Fs in pulp mill effluent in the past were the chlorination of dibenzo-*p*-dioxin precursors during bleaching and the condensation of polychlorinated phenols during pulp digestion (Luthe and Prahacs 1993; Luthe *et al.* 1993). The elimination of dibenzo-*p*-dioxin precursors from defoamer products, exclusion of chlorophenol-contaminated wood chips and introduction of chlorine dioxide bleaching have dramatically reduced mill-related contamination (Whitehead *et al.* 1992; Luthe *et al.* 1992). The introduction of PCDD/Fs from combustion (both from the mill and ambient deposition) into mill effluent is a more minor process, but has proved to be more difficult to eliminate (Hites 1990; Luthe and Prahacs 1993).

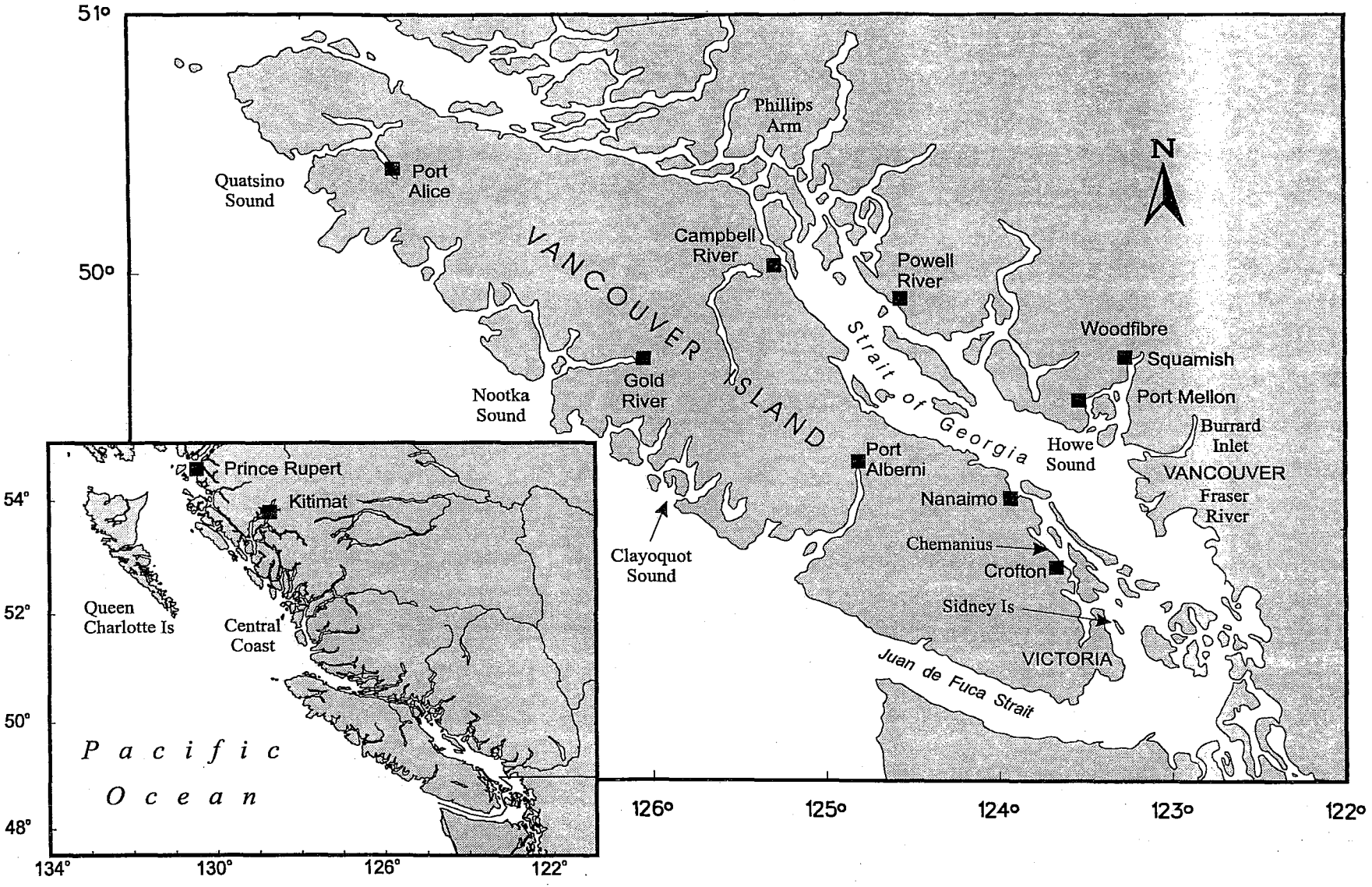
Monitoring programs were initiated in 1989 to document the sources and concentrations of PCDD/Fs in B.C. harbours and pulp mill sites. These programs were designed to assess ecosystem impact, address human health concerns due to contamination of the fishery and assist in the formulation of regulations for fisheries management. Aspects such as the geographical extent of contamination, persistence of PCDD/Fs over time, and trends in the congener composition and proportion of toxic congeners were also important considerations. However, translation of these aspects into monitoring programs has produced a very large data set. For crab hepatopancreas alone research and monitoring activities since the late 1980s in the vicinity of 10 coastal mills and the principal harbours (Table 1-1; Map 1-1) had produced chlorinated dibenzo-*p*-dioxin and dibenzofuran concentrations for nearly 500 samples. A data set this size can only be effectively interpreted with multivariate analysis techniques.

In this work we employ Principal Components Analysis (PCA) to explore the interrelationships and trends in both the chlorinated dibenzo-*p*-dioxin and dibenzofuran patterns and the toxic congener composition among B.C. mill sites and between harbours and mill sites. The integration of toxic-equivalency into the interpretation provides a mechanism for addressing both ecosystem effects and potential health risks to humans. An abbreviated account of this work has appeared in Yunker and Cretney (1996).

Table 1-1. Coastal B.C. sampling sites, their physical environment and the pulping process used by the different mills.

Site	Geographical Location	Physical Environment	Primary Mill Process
Pulp mill sites			
Port Alberni	Fjord	Restricted circulation	Bleached Kraft
Crofton	Channel	Well flushed	Bleached Kraft
Campbell River (Elk Falls Mill)	Channel	Well flushed	Bleached Kraft
Gold River	Fjord	Restricted circulation	Bleached Kraft
Howe Sound (Port Mellon and Woodfibre Mills)	Fjord	Restricted circulation	Bleached Kraft
Kitimat [a]	Fjord	Restricted circulation	Unbleached Kraft
Nanaimo (Harmac Mill)	Channel	Well flushed	Bleached Kraft
Powell River	Strait	Well flushed	Bleached Kraft
Prince Rupert	Narrow Channel	Restricted circulation	Bleached Kraft
Harbours and industrialized areas			
Vancouver Harbour (Burrard Inlet)	Fjord	Restricted circulation	
Nanaimo Harbour	Bay	Restricted circulation	
Victoria/Esquimalt Harbours	Bay	Restricted circulation	
Fraser River mouth	Estuary	Well flushed	

[a] Kitimat is also a major harbour and is the site of an aluminum smelter and a methanol production plant (see Section 13).



Map 1-1. Map of the British Columbia coast showing the pulp mills and the principal harbours.

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2. PCA MODEL BUILDING

Data for the concentrations of chlorinated dibenzo-*p*-dioxins and dibenzofurans in 469 samples of crab hepatopancreas were obtained primarily in electronic form from the Federal Departments of Fisheries and Oceans and Environment. Samples from a few mill sites from 1988 and 1989 were not analyzed for the full suite of congeners and could not be included in the data set. All data were then reverified against original laboratory reports. Data were primarily for Dungeness crab (*Cancer magister*) hepatopancreas, although 13 hepatopancreas samples from Red Rock crabs (*Cancer productus*), six from Box crabs (*Lopholithodes spp*) and one sample from a Kelp crab (*Pugettia producta*) were also included.

The data set included 11 benchmark samples from 1987 (i.e. prior to any mill process changes) which were obtained from a Canadian Wildlife Service unpublished report (Norstrom *et al.* 1988). Post 1987 monitoring data for the mill sites included 378 samples analyzed by Axys Analytical Ltd. of Sidney B.C. (formerly Seakem Analytical Ltd.), 15 samples analyzed by Envirotest Ltd. of Edmonton Alberta and eight samples analyzed by Wellington Laboratories Ltd. of Guelph Ontario. The 1989 and 1993 samples from Kitimat and Nanaimo respectively were analyzed by Envirotest and the 1993 samples from Gold River were analyzed by Wellington Laboratories. Data for samples from remote locations have principally been produced by the Department of Fisheries and Oceans, with 30 samples from the Great Lakes Laboratory for Fisheries and Aquatic Sciences in Burlington Ontario and 25 samples from the Institute of Ocean Sciences in Sidney B.C.

Consistent collection, dissection, extraction and analysis procedures have been maintained for the crab samples analyzed by Axys Analytical (more than 80% of the data set). All samples were collected and dissected by Hatfield Consultants and were all subjected to the same extraction protocol by Axys Analytical (e.g., Dwernychuk 1993). All samples were spiked with ^{13}C-labelled surrogate standards for the 2,3,7,8-tetra- to octachlorodibenzo-*p*-dioxins plus 2,3,7,8-tetrachlorodibenzofuran; as a minimum ^{13}C-labelled 1,2,3,4-tetrachlorodibenzo-*p*-dioxin was added as a recovery standard. For samples analyzed by high resolution GC/MS (i.e. 1991 onwards), ^{13}C-labelled 2,3,7,8-penta- to heptachlorodibenzofuran surrogates and 1,2,3,7,8,9-hexachlorodibenzo-*p*-dioxin and 1,2,3,4,6,7,8-heptachlorodibenzo-*p*-dioxin recovery standards were also added. Minor method differences occurred with the other laboratories, but all analyses employed ^{13}C-labelled standards.

Unsupervised principal components analysis was performed using the nonlinear iterative partial least squares (NIPALS) algorithm in a program which allowed a detailed examination of both crab sample and variable (congener) projections (respectively scores and loadings; software was provided by the Chemometrics Clinic, Seattle WA). Congeners and congener totals which were undetected (ND) were replaced by the limit of detection. Values with questionable ion ratios (NDR) were used only if the concentration was reasonable in the context of similar samples from the site.

For most PCA variables the limit of detection was used to estimate the concentration for less than 15% of the samples (Table 2-1). The more chlorinated congeners, particularly OCDD, OCDF and the HpCDFs, were undetectable in higher percentages of samples and were used only when it was verified that their presence did not skew the model. The congeners 1,2,3,4,7,8-HxCDD and 1,2,3,4,7,8,9-HpCDF were undetectable in most samples and were removed. Because the 2,3,7,8-substituted HxCDFs were only detectable in a few samples, an average of the detection limits or concentrations for the individual congeners was used. For each congener series a non-2,3,7,8 congener total was obtained by subtracting the 2,3,7,8-substituted congener(s) from the homologue total; if a value of zero resulted for these "other" dioxins, the detection limit for the homologue total was substituted.

Normalization of each sample was employed before PCA, because: 1) concentrations differed by orders of magnitude, particularly between years and 2) a primary goal was to examine trends in the chlorinated dibenzo-*p*-dioxin and dibenzofuran congeners with a "mill" pattern. Log transformation was also evaluated, but the resulting PCA model was more sensitive to detection limit artifacts. Negative correlations due to closure of the largest variables were avoided during normalization by using a subset of the variables with mid-range standard deviations and means (Johansson *et al.* 1984; Yunker *et al.* 1995). The subset of congeners used to calculate normalization factors for each sample is listed in Table 2-1. Data were then autoscaled (mean centered and divided by the variance on a variable-by-variable basis) before PCA.

The use of the limit of detection as an estimation of the actual concentration proved to be a problem for 18 low concentration samples that either were from remote areas or were earlier (1989-91) samples with high detection limits (Table 2-2). These samples had only 2-6 congeners detectable out of 20 (average 4), and the limit of detection (and hence apparent concentration) was usually noticeably higher for the higher chlorine-numbered congeners (e.g., Figure 2-1 to Figure 2-3). The resulting dominance of hepta- and octachlorinated congeners produced a spurious separation of these samples from all other samples in the first principal component (PC). When these samples were excluded the hepta- and octachlorinated congeners became much less important to the PCA model (see following). To simplify source interpretation a Varimax axis rotation was then applied to the first three PCs.

Table 2-1. PCA model abbreviations, percent of samples for which the congeners used in the PCA model were undetectable, 2,3,7,8-toxic equivalent factors for the PCA congeners and the variables used to calculate a normalization factor.

Congener	PCA Abbrev.	Percent Undetectable	TEF	Normalizing Variables
2,3,7,8-TCDD	4D	8.1	1.0	4D
Non-2,3,7,8 TCDD	4DO	5.8	0	
1,2,3,7,8-PnCDD	5D	11.1	0.5	5D
Non-2,3,7,8 PnCDD	5DO	9.2	0	5DO
1,2,3,6,7,8-HxCDD	6D2	1.1	0.1	
1,2,3,7,8,9-HxCDD	6D3	12.2	0.1	6D3
Non-2,3,7,8 HxCDD	6DO	0.4	0	
1,2,3,4,6,7,8-HpCDD	7D	15.4	0.01	7D
Non-2,3,7,8 HpCDD	7DO	12.8	0	7DO
OCDD	8D	39.2	0.001	8D
2,3,7,8-TCDF	4F	0.6	0.1	
Non-2,3,7,8 TCDF	4FO	0.6	0	
1,2,3,7,8-PnCDF	5F1	29.9	0.05	5F1
2,3,4,7,8-PnCDF	5F2	14.1	0.5	5F2
Non-2,3,7,8 PnCDF	5FO	7.2	0	5FO
2,3,7,8-HxCDF mean	6F	66.3	0.1	
Non-2,3,7,8 HxCDF	6FO	23.0	0	6FO
1,2,3,4,6,7,8-HpCDF	7F1	41.2	0.01	7F1
Non-2,3,7,8 HpCDF	7FO	39.4	0	
OCDF	8F	87.2	0.001	

Table 2-2. Samples with too few congeners detectable to be included in the PCA data set.

Area/Year	Sample Date	Site Number	Location	Species	Congeners Detectable	Calculated TEQ pg/g
Queen Charlotte Is. 91	19-Mar-91		Government Creek	Dungeness Crab	4	0.2
Central Coast 91	19-Mar-91		Burke Channel	Dungeness Crab	4	0.2
Crofton 90	14-Aug-90	C14	Satellite Channel	Dungeness Crab	4	2.0
Crofton 90	16-Aug-90	C15	Satellite Channel	Dungeness Crab	4	7.1
Crofton 90	20-Aug-90	C16	Houston Channel	Dungeness Crab	6	7.6
Crofton 90	20-Aug-90	C16A	Houston Channel	Dungeness Crab	6	21.1
Crofton 90	15-Aug-90	C19	Saanich Inlet	Dungeness Crab	4	4.9
Crofton 90	16-Aug-90	C21	Saanich Peninsula - North	Red Rock Crab	4	1.7
Nanaimo 90	24-Aug-90	C10	Hammond Bay	Dungeness Crab	4	4.4
Port Alberni 90	04-Jul-90	C6	Sproat Narrows	Dungeness Crab	5	3.1
Powell R. 90	07-Sep-90	C7	Savary Is. - North	Dungeness Crab	4	9.4
Powell R. 90	04-Sep-90	C8	Savary Is. - South	Red Rock Crab	2	0.9
Powell R. 90	05-Sep-90	C11	Algerine Passage	Dungeness Crab	4	5.1
Prince Rupert 90	06-Jun-90	SC1A	Morse Basin	Dungeness Crab	5	16.6
Prince Rupert 91	22-Mar-91	SC4	Kitson Island - West	Dungeness Crab	6	7.9
Prince Rupert 91	22-Mar-91	SC5	Coast Island - South	Dungeness Crab	6	6.9
Prince Rupert 91	22-Mar-91	SC6	Kinahan Islands	Dungeness Crab	4	1.5
Kitimat Arm 89	16-Dec-89	S4	Gobeil Bay	Dungeness Crab	2	0.8

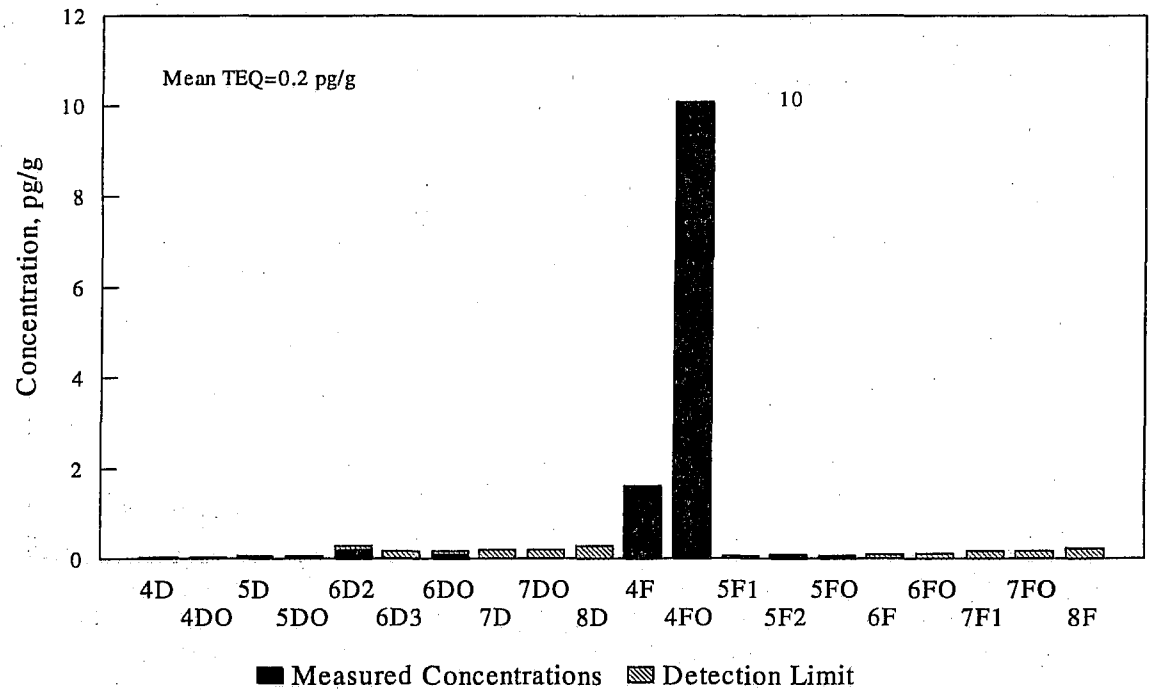

Figure 2-1. Samples from the Central Coast and Queen Charlotte Islands in 1991 (n=2).

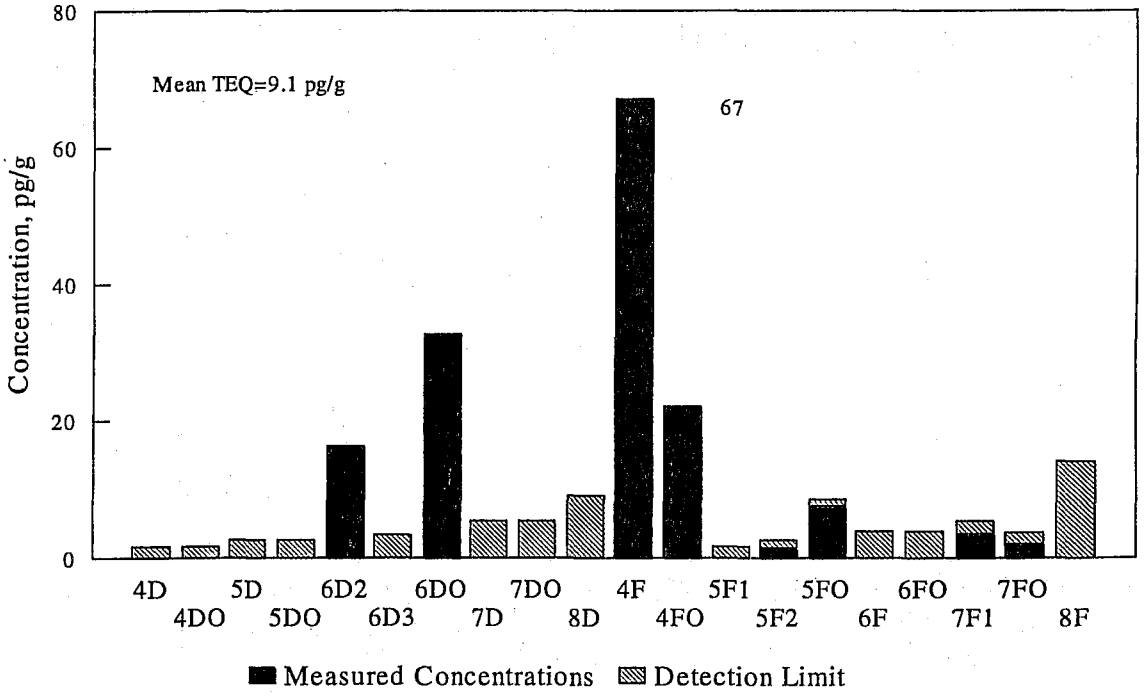

Figure 2-2. Samples C10 (1990) and C1 (1991) from the Nanaimo area (n=2).

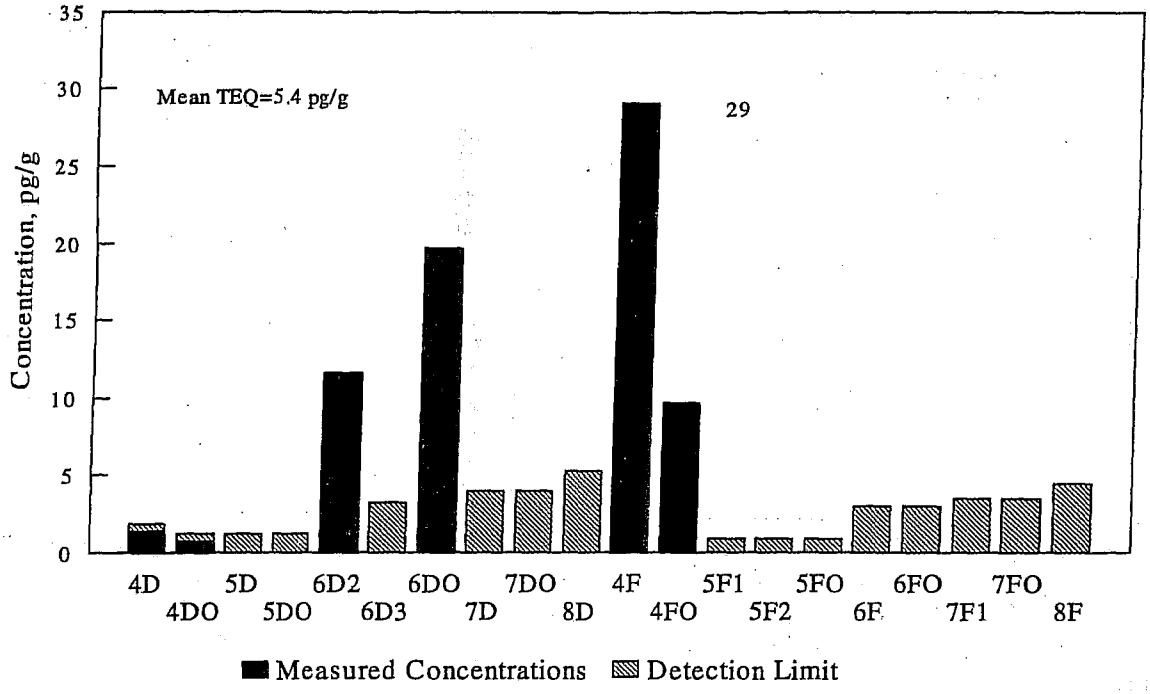

Figure 2-3. Prince Rupert (1991) samples SC4, SC5 and SC6 (n=3).

3. OVERVIEW OF PCA RESULTS

The utility of this data set for the interpretation of trends in congener composition both between sites and over time is greatly enhanced by having more than 80% of the samples analyzed by one laboratory. Furthermore, the normalization of samples before PCA removed potential concentration biases and allowed samples from different laboratories to be added to the model. Concentration profiles and PCA projections for the non-Axys Analytical samples were entirely consistent with the Axys samples from similar sites or the same site in different years.

THE PCA MODEL

PCA is invaluable for classifying chlorinated dibenzo-*p*-dioxin and dibenzofuran congener variables according to their primary source (e.g., chlorine bleaching, pentachlorophenol) and for identifying congeners that covary. PCA separates a data matrix into the product of two matrices (one each for samples and variables) with the PCs comprising individual column or row vectors in their respective matrix. Each PC provides interrelated information concerning trends in the samples and the variables. The PCA model is calculated with no *a priori* assumptions about the data structure and completely unbiased projections result. A very readable explanation of the application of PCA to chemical data has been published by Meglen (1992).

The crab hepatopancreas PCA model used throughout this report includes 451 samples and employs 20 PCDD/F variables. Before Varimax axis rotation the variance accounted for by the model in respectively the first, second and third PCs was 27.4%, 17.4% and 14.6%, for a total of 59.4%. The first five PCs all had eigenvalues greater than 1.0, and thus represented real (as opposed to random) variation in the data set (Meglen, 1992). After axis rotation of the first three PCs the variance in the three PCs was redistributed to 22.1%, 18.8% and 18.5%.

Variable and sample projections for the PCA model are given in Figure 3-1 and Figure 3-2. General features of the model are discussed in this section. In the area by area discussion that follows PCA sample plots are presented that show subsets of the larger model to illustrate, for example, the trends in samples from a given location in a given year. Hence these subset sample plots are only visual simplification not a new PCA model based on a smaller number of samples, and the general discussion in this section applies to every PCA plot shown in this report.

In Figure 3-1 the PCDD/F variables project together according to source. Congeners that are produced primarily by chlorination of dibenzo-*p*-dioxin precursors during pulp mill chlorine bleaching project to the far right and include 2,3,7,8-TCDF, the non-2,3,7,8 TCDFs and the two 2,3,7,8-PnCDF isomers (e.g., Luthe and Prahacs 1993). Dibenzo-*p*-dioxins that are produced by the condensation of polychlorinated phenols during pulp digestion cluster in the bottom of Figure 3-1, close to the *y*-axis, and include 1,2,3,7,8-PnCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD and the non-2,3,7,8 HxCDD isomers (Luthe *et al.* 1993). 2,3,7,8-TCDD projects midway between these two groups, which suggests mixed sources. Congeners that are present in

pentachlorophenol wood preservative but are removed during the pulping process project at the top of Figure 3-1 and include 1,2,3,4,6,7,8-HpCDF, the non-2,3,7,8 TCDFs and HxCDFs, and to a lesser extent the non-2,3,7,8 HpCDFs (Hagenmaier and Brunner 1987; Luthe *et al.* 1993). With the exception of 1,2,3,4,6,7,8-HpCDF, all of the above congeners have significant loadings only in the first two PCs.

Congeners at or near the limit of detection (Table 2-1), such as 2,3,7,8-HxCDF, OCDF, OCDD and the HpCDDs project close to the origin in the first two PCs but make a strong contribution to the third PC. Thus the contributions of detection limit artifacts and/or combustion have been largely shifted to the third PC. The remaining variable, non-2,3,7,8 PnCDD, makes a contribution to all three PCs and is apparently present in all sample types.

The projection of the toxic 2,3,7,8-substituted TCDD, TCDF and PnCDDs on the right side of Figure 3-1 produces a correlation between samples and their toxicity in Figure 3-2. The log of the 2,3,7,8-TCDD toxic equivalents (TEQ) is highly correlated (r^2=0.42; p<0.001) with the projection of each sample in the first PC (t1). The TEQ correlation with the second PC (t2) is lower but is still significant (r^2=0.17; p<0.001). The correlation of the log TEQ with t1 and t2 is only slightly improved over the correlation with t1 (r^2=0.44; p<0.001).

There is a direct source correspondence between the positions of the variables in Figure 3-1 and the positions of the samples in Figure 3-2. In Figure 3-2 samples with similar distributions of the 20 PCDD/F congeners and congener totals cluster or project together. Samples that are far apart have a different "fingerprint" of the 20 congeners and the crabs collected for these samples have been exposed to a different suite of PCDD/Fs.

Crab samples projecting to the far right in Figure 3-2 have the highest proportion of congeners characteristic of pulp mill chlorine bleaching, while samples in the lower centre have been most strongly influenced by polychlorinated phenol condensation. Samples in between these two areas have been exposed to varying amounts of these two inputs.

Crab samples projecting in the upper left of Figure 3-2 have been exposed to pentachlorophenol wood preservatives and are from the major B.C. harbours. These samples have low proportions of the toxic congeners, but have high proportions of the non-2,3,7,8 congeners — principally the PnCDFs and HxCDFs and the HxCDDs. Samples collected in different years project together indicating little or no change in composition over time.

Crabs do not appear to take up hepta- and octachloro dibenzo-*p*-dioxins and dibenzofurans to any great extent: these congeners were generally only detectable when concentrations were very high. One important consequence is that the contribution of atmospheric combustion (the typical pattern is predominant OCDD with lesser amounts of HpCDD and HpCDF; Hites 1990) can not be evaluated based solely on crab data.

APPLICATION OF PCA TO B.C. COASTAL SITES

Crabs from the vicinity of nine B.C. coastal Kraft mills at eight sites have been exposed to chlorine bleach plant effluents (Table 1-1). The tenth mill at Kitimat does not use chlorine bleaching. Crabs from the principal harbours have all been exposed to chlorophenols from wood treatment facilities as well as to PCDD/Fs from combustion. Crabs from reference locations, which include Sidney Island (north of Victoria), the Queen Charlotte Islands and the B.C. Central Coast, were expected to have received minimal exposure to PCDD/Fs. Each area is discussed individually in the sections that follow.

Averaged congener profiles for crab samples with similar PCA projections that are from locations geographically close to each mill site provide a direct connection between congener composition and the PCA results. Congener profiles are presented for the earliest and most recent sampling times in Figure 3-3 and Figure 3-4; the specific samples used for each profile are tabulated in the individual sections. In each case the tip of the arrow indicates the mean projection of the samples that have been used to define each profile. In all cases where 1987 and 1989/90 samples have been averaged there was little or no change in either composition profile or PCA projection over the time interval shown. For Campbell River (Elk Falls) and Powell River the crab sample collected closest to the mill site sometimes had a PCA projection similar to that of the harbours (see Sections 7 and 11). Because this likely indicates the presence of pentachlorophenol wood preservatives, these samples were not included in the profiles.

Examination of Figure 3-3 and Figure 3-4 reveals striking differences between the chlorinated dibenzo-*p*-dioxin and dibenzofuran profiles of different mill sites. At Gold River, for example, the 1990 profile is made up almost entirely of TCDFs. In contrast, the 1989 profile from Kitimat shows a pattern consistent with polychlorinated phenol condensation, without any inputs from chlorine bleaching. This condensation was likely a result of heating pentachlorophenol contaminated wood chips during the Kraft pulping process (Luthe *et al.* 1993). In 1987/90 locations such as Nanaimo (Harmac Mill), Port Alberni and Campbell River contained equal proportions of the TCDFs and the non-2,3,7,8 HxCDDs. The mill at Crofton had a lower proportion of the HxCDDs and the mills at Powell River, Howe Sound and Prince Rupert were similar to the Gold River mill in having only small proportions of the HxCDDs. The composition of Gold River samples indicates either that very few pentachlorophenol contaminated chips were used in the bleach plant, or less likely, that digester conditions did not favour HxCDD formation.

Over the four or five years since the mills introduced changes to control the discharge of PCDD/Fs, the TCDFs have been removed faster than the HxCDDs at the mill sites, while the harbour profiles have shown little change over time (Figure 3-4). At each mill site an increase in the proportion of the HxCDDs has been accompanied by a corresponding decrease in the TCDFs. While the mill sites have maintained much of their individual character, the overall effect has been for the composition profiles and PCA projections to become more similar over time.

Examination of profile and PCA results for the Nanaimo area illustrates that different PCDD/F fingerprints can be obtained for locations which are only ca. 5 km apart (Figure 3-3 and Figure 3-

4; Section 9). In general crab samples from the Harmac mill site project with similar mill samples (close to the *y*-axis), while samples from Nanaimo Harbour project close to the Victoria and Vancouver Harbour samples. Nanaimo Harbour samples have a slightly higher proportion of the TCDFs than the larger harbours, and some contribution of chlorine bleaching effluents from the mill is likely. At a few locations from the boundary area just outside Nanaimo Harbour, crab samples had a mill pattern one year and a harbour pattern in another year — this highlights the utility of PCA in assigning a contaminant source to individual crab samples.

In general 1987 samples had the highest concentrations of toxic congeners (Table 3-1). In subsequent years both lower proportions of the toxic 2,3,7,8-chlorinated congeners and a decrease in overall PCDD/F concentrations have been observed. The result has been a decrease in the TEQ at B.C. mill sites.

Mills located on a fjord (where the bottom water exchanges very slowly) or in an area with restricted circulation had the highest proportion of toxic congeners in the earliest years of sampling, but have shown the largest TEQ reduction over time (Table 1-1 and Table 3-1). The Howe Sound and Gold River mills provide good examples. In quiescent areas burial plays an important role in the removal of the particulate associated dioxins and furans (cf. Macdonald *et al.* 1992). Within Howe Sound the reduction in TEQ has been more rapid adjacent to the Woodfibre mill; here the large amount of sediment delivered by the Squamish River is the most likely reason for the difference. Hence, the amount of sediment available for burial at each mill site is an important factor in contaminant removal.

In contrast, samples from locations which are well flushed by tidal currents had lower initial proportions of the toxic congeners than samples from inlets but have exhibited a smaller decrease in TEQ over time. The Crofton and Campbell River locations illustrate this trend. In this case burial rates are either too low or resuspension processes are too active for effective removal of contaminants by burial. The high current rates off Campbell River facilitate sediment transport and dispersal may play a more important role in contaminant removal in this area.

The principal harbours have been less intensively studied, but they would appear to present a different situation. These locations are adjacent to metropolitan areas, with the potential for chronic inputs. Furthermore sedimentation and burial rates are likely to be lower in harbours than they are in fjords that are fed by a major river (e.g., the Squamish River in Howe Sound).

It has proven to be difficult to define a background pattern for crabs on the B.C. coast. Earlier samples from remote areas were analyzed by low resolution mass spectrometry, with too few congeners detectable to provide a reliable PCA projection. Subsequent samples have generally been collected from locations away from the mill monitoring areas, but they may have been impacted by mill discharges (see Section 4). However, even samples from areas more removed from mills and harbours have PCA projections and composition profiles different from the usual atmospheric pattern (cf. Hites 1990). This topic requires further study.

COMPARISON OF DIFFERENT CRAB SPECIES

The different crab species that are included in this data set feed in different ecological niches. Dungeness crabs are bottom scavengers; their diet consists primarily of clams, but they also feed on smaller crustaceans and fish and will feed opportunistically on fish carcasses (Jensen, 1995). Although smaller in size, Red Rock crabs have a similar diet, but tend to frequent rocky habitats in shallower water, and are likely to scavenge on smaller animals. Box crabs feed on sea urchins and other echinoderms; they are one of the few crab species that feed on anemones. Kelp crabs are situated further down the food chain; they feed mostly on vegetation, but will feed opportunistically on jellyfish. These differences in diet, and in some cases trophic level, result in exposure to different concentrations and proportions of PCDD/Fs for the different crabs.

To test for a relationship between species and PCDD/F congener composition, the PCA projections of Red Rock and Box crabs are compared to the projections of Dungeness crabs that were collected at the same time from the same environment (i.e. areas at a similar distance from the mill, with the same circulation pattern, etc.). In each case arrows are used to highlight the difference in PCA projection relative to the most suitable Dungeness crab sample (Figure 3-5 and Figure 3-6; Table 3-2). For a number of samples (e.g., Red Rock crabs from Lasqueti Island or a number of the Box crabs from around Quadra Island; Sections 5 and 7) no concurrent samples are available. Dungeness crab samples are also not available for comparison to the one sample of Kelp crab (Section 7).

PCA projections of the Red Rock crab samples shift in a number of different directions relative to the Dungeness crab samples, which suggests an overall similarity in the congener compositions (Figure 3-5). However, on balance the Red Rock crabs appear to have a lower proportion of the most toxic congeners. In agreement, congener totals and calculated TEQs are consistently lower for samples of Red Rock crab (Table 3-2). Fewer sample pairs are available for the Box crabs, but the Box crabs consistently project to the lower left of the Dungeness crabs, and in most cases have a lower congener total and TEQ (Figure 3-6; Table 3-2). While these differences in composition may in part be due to differences in sampling location and hence the amount of exposure to mill effluent experienced by the crabs, the PCA results suggest that differences in feeding behavior have resulted in a reduction in the toxic load for both of the non-Dungeness species.

Given these interspecies differences in PCA projections and TEQs, it is evident that whenever possible a single species — Dungeness crab — should be used for the mill monitoring programs. The use of a single species is also preferable for statistical evaluation of trends. In addition, because the Dungeness crabs apparently ingest among the highest proportions of toxic congeners of any crab species, they provide a useful and highly defensible indicator for both fishery closures and openings.

Table 3-1. Dungeness crab hepatopancreas congener total and TEQ averages obtained using samples with similar PCA projections.

Sampling Location	1987			1989/90			1992/94		
	Congener Total [a]	Calculated TEQ [a]	n [b]	Congener Total [a]	Calculated TEQ [a]	n [b]	Congener Total [a]	Calculated TEQ [a]	n [b]
	pg/g wet weight			pg/g wet weight			pg/g wet weight		
Principal Harbours	1700	77	3	-	-	-	770	23	17
Nanaimo Harbour	470	27	1	300	15	2	360	18	4
Port Alberni	-	-	-	450	23	6	230	11	4
Crofton	4300	420	2	1700	130	5	1500	94	3
Campbell River (Elk Falls)	-	-	-	1200	100	3	800	53	3
Gold River	-	-	-	13,000	1100	5	1200	99	4
Howe Sound	30,000	2900	2	9900	850	6	1000	95	8
Kitimat	-	-	-	570	68	8	110	7.2	41
Nanaimo (Harmac) Mill	1100	82	1	810	65	5	290	14	3
Powell River	-	-	-	620	57	5	160	12	2
Prince Rupert	18,000	1900	1	2900	260	3	1000	53	4

[a] The specific samples used for these calculations are detailed in tables that are included in each of the individual sections.
[b] n is the number of samples.

Table 3-2. Congener totals and calculated TEQs for the samples of Dungeness, Red Rock and Box crab used in the species comparison.

Sample/Year	Station	Location [a]	Congener Total	Calculated TEQ	Station	Location [a]	Congener Total	Calculated TEQ
			pg/g				pg/g	
	Dungeness Crab				*Red Rock Crab*			
Campbell R. 90	C5	Gowlland Harbour	1800	130	C2	May Island	210	21
Nanaimo 90	C5	Pylades Channel	440	44	C4	Northumberland Ch.	99	6.8
Nanaimo 90	C6	Ruxton Is.	210	20	C7	Stuart Channel	200	11
Nanaimo 91	C4	North Mudge Is.	480	36	C4	North Mudge Is.	170	14
Nanaimo 91	C4	North Mudge Is.	460	35				
Sechelt 93 [b]	C303	Secret Cove	220	10	C302	Secret Cove	33	1.4
Powell R. 93	C16	Hernando Is.	120	9.6	C15	Copeland Is.	30	2.2
	Dungeness Crab				*Box Crab*			
Powell R. 90	C2	Harwood Is.	690	64	C3	Scuttle Bay	1600	140
Campbell R. 92	C15	Marina Island	320	25	C9	Sutil Channel	180	14
Howe Sound 93	C21	Gower Point	670	45	C23	Offshore Gower Pt.	260	16
Howe Sound 93	C21	Gower Point	690	47				

[a] Station locations are given on maps in each individual section.
[b] See Map 5-2.

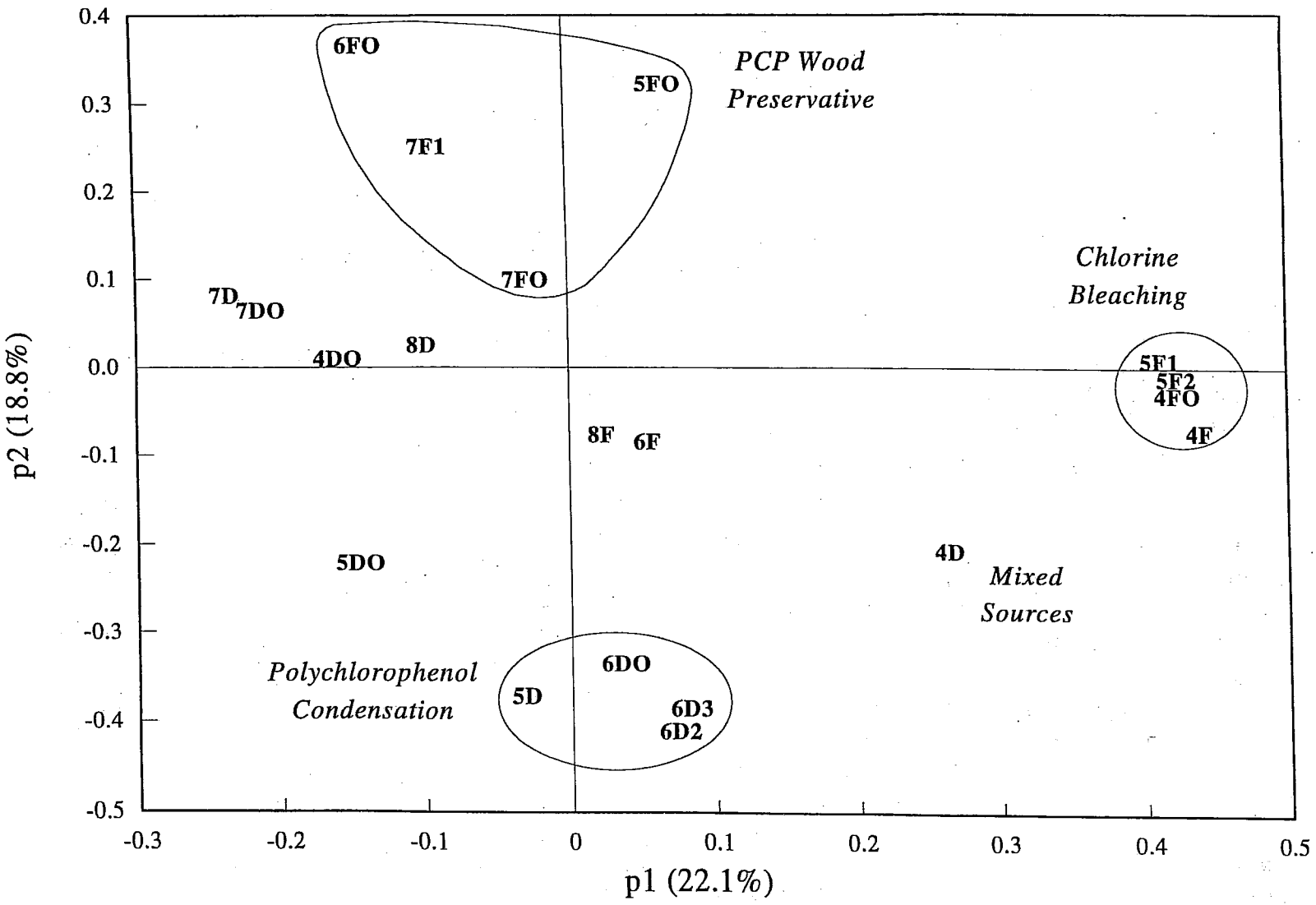

Figure 3-1. Varimax rotated variable (loadings) plot for PCDD/F congener projections in the first and second PCs.

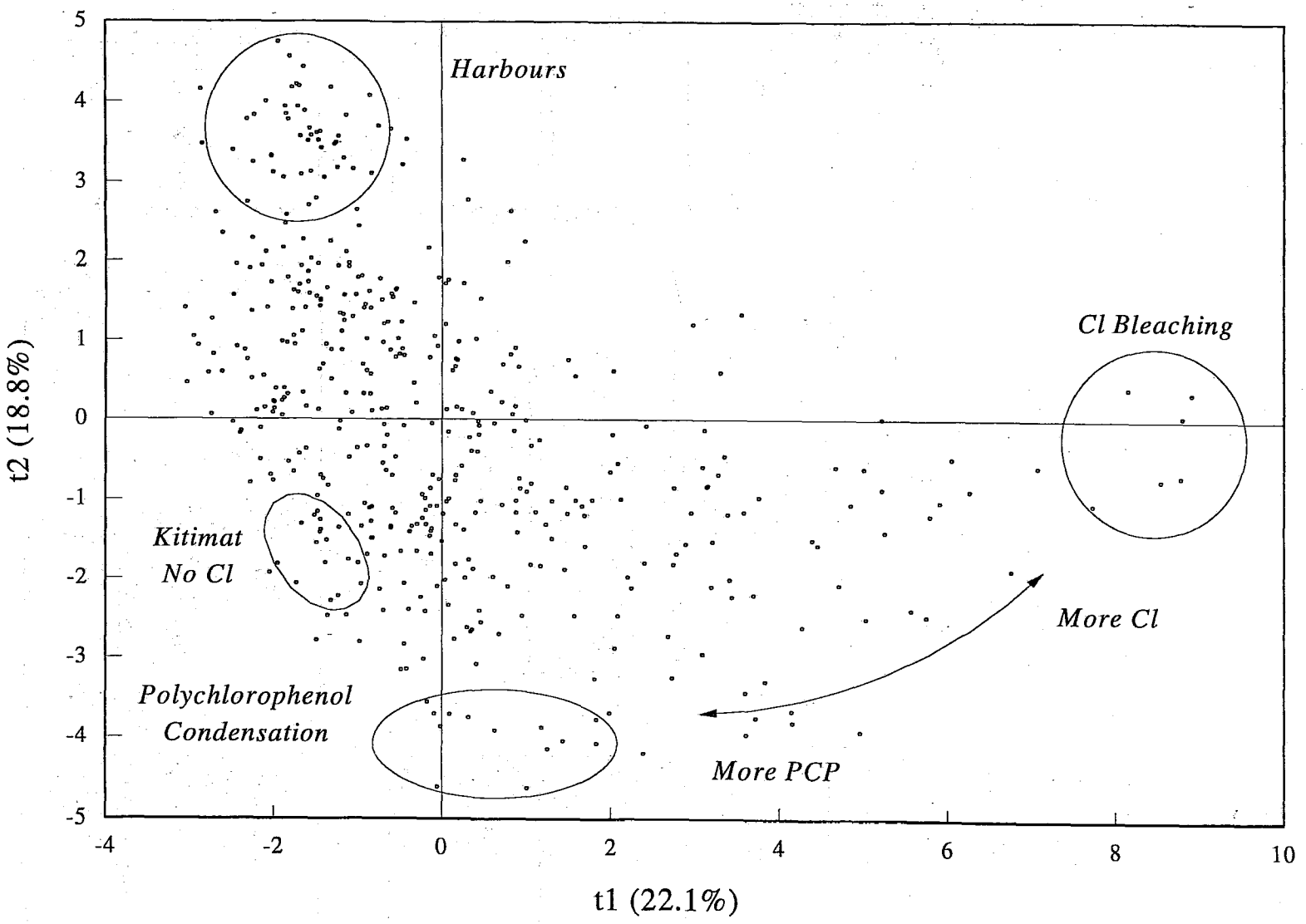

Figure 3-2. Varimax rotated sample (scores) plot for crab hepatopancreas samples in the first and second PCs.

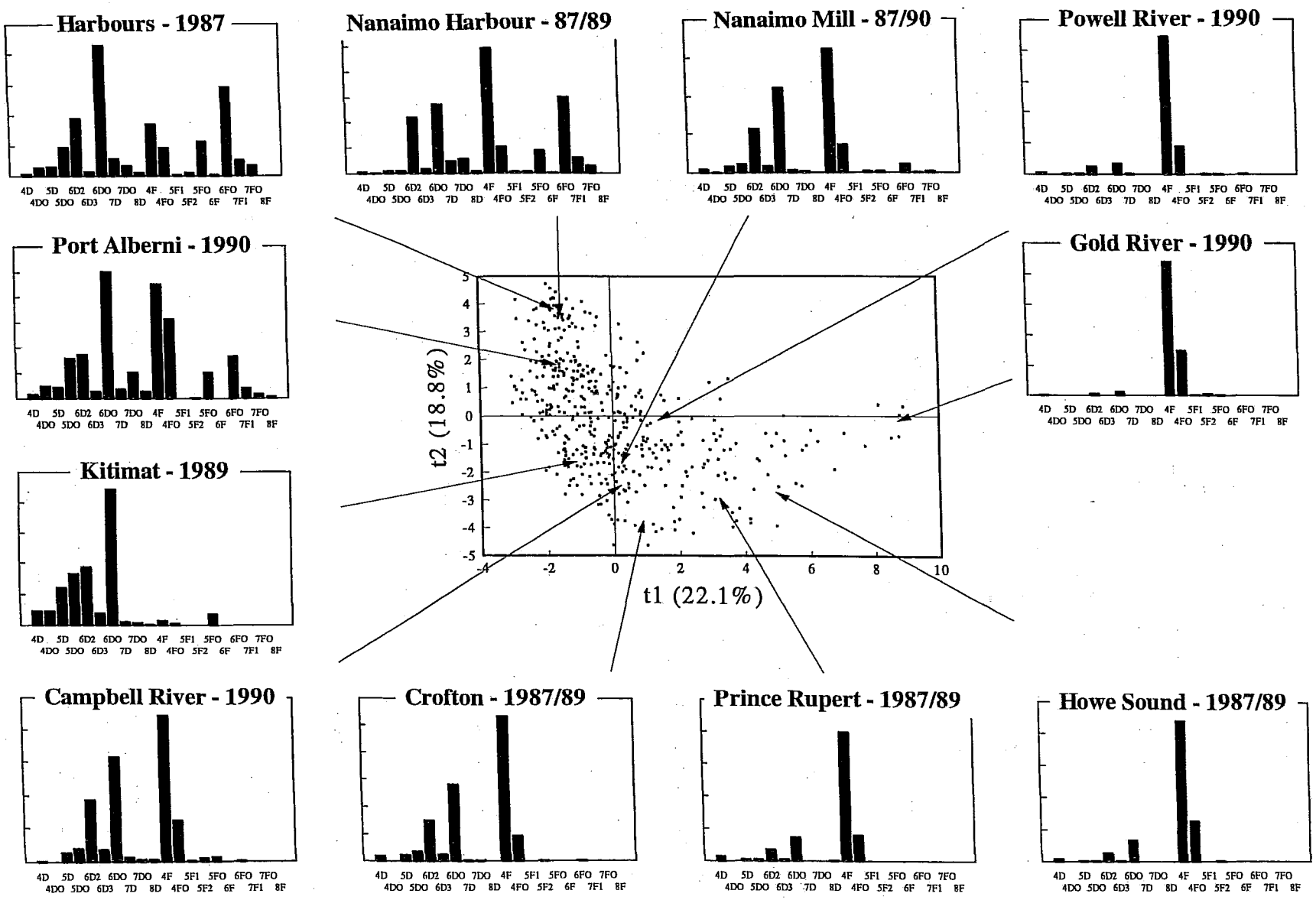

Figure 3-3. Crab sample congener profiles and PCA projections for the earliest sampling times available.

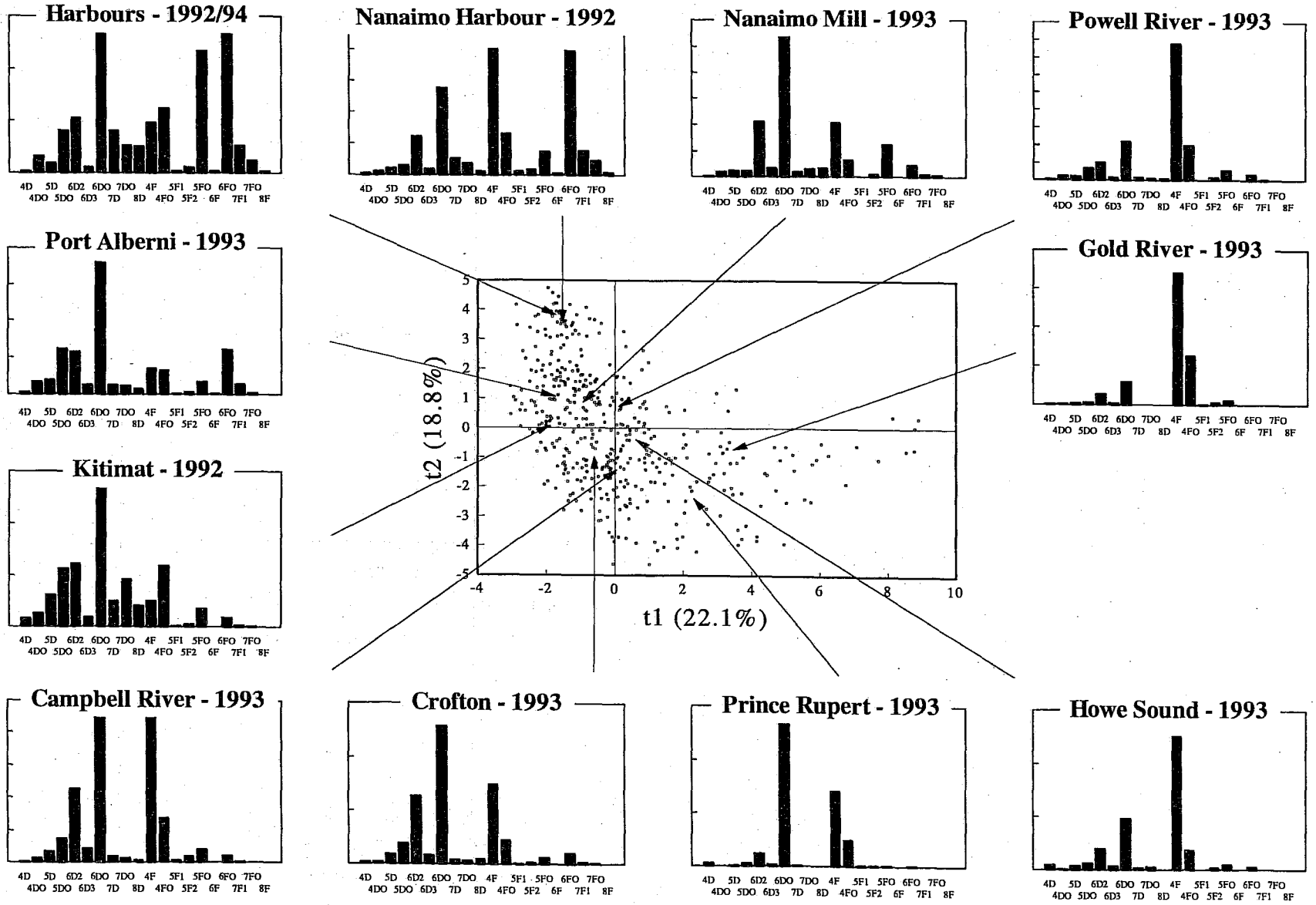

Figure 3-4. Crab sample congener profiles and PCA projections for the most recent sampling times.

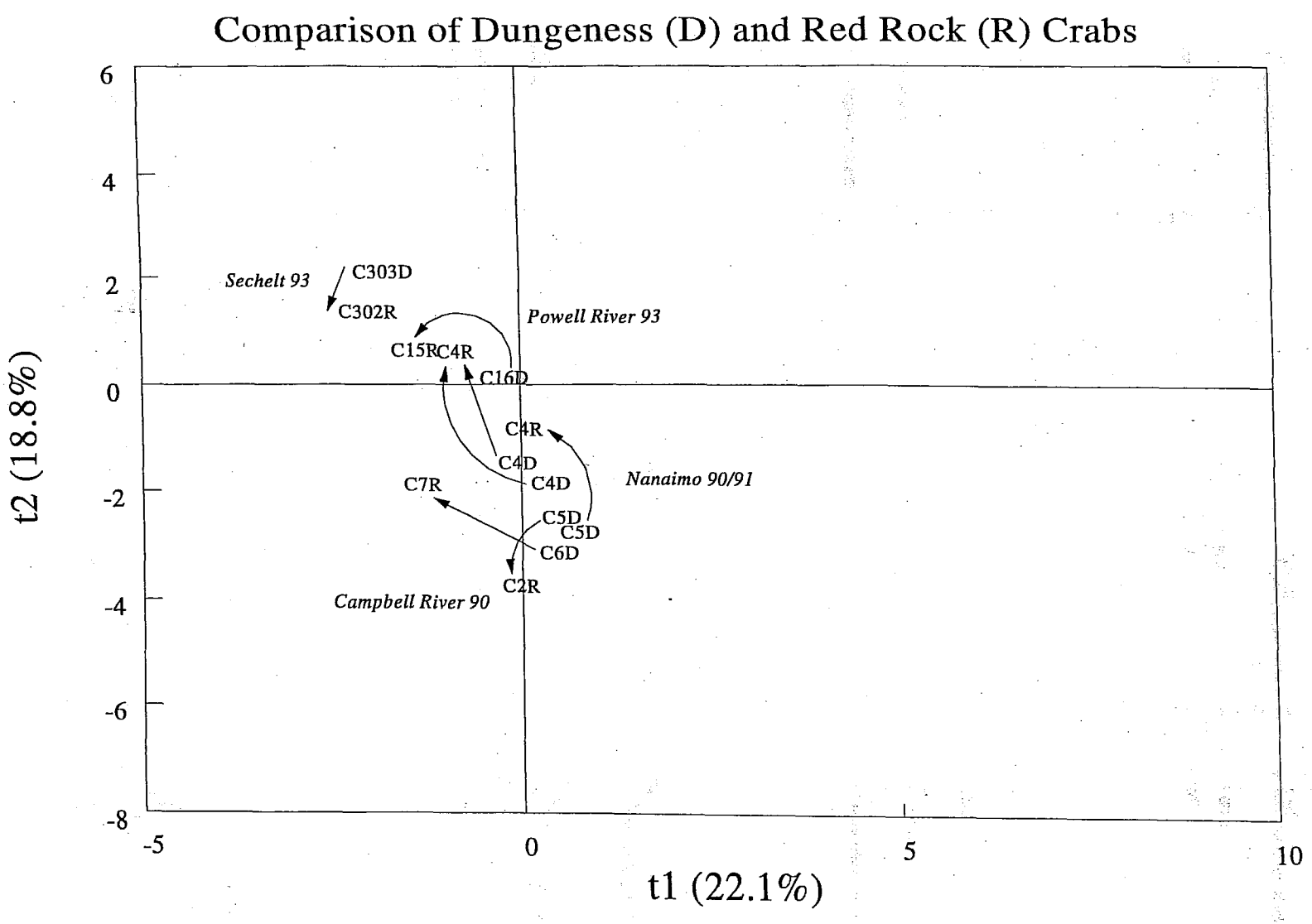

Figure 3-5. Dungeness and Red Rock crab samples from Nanaimo, the Sechelt peninsula, Campbell River and Powell River.

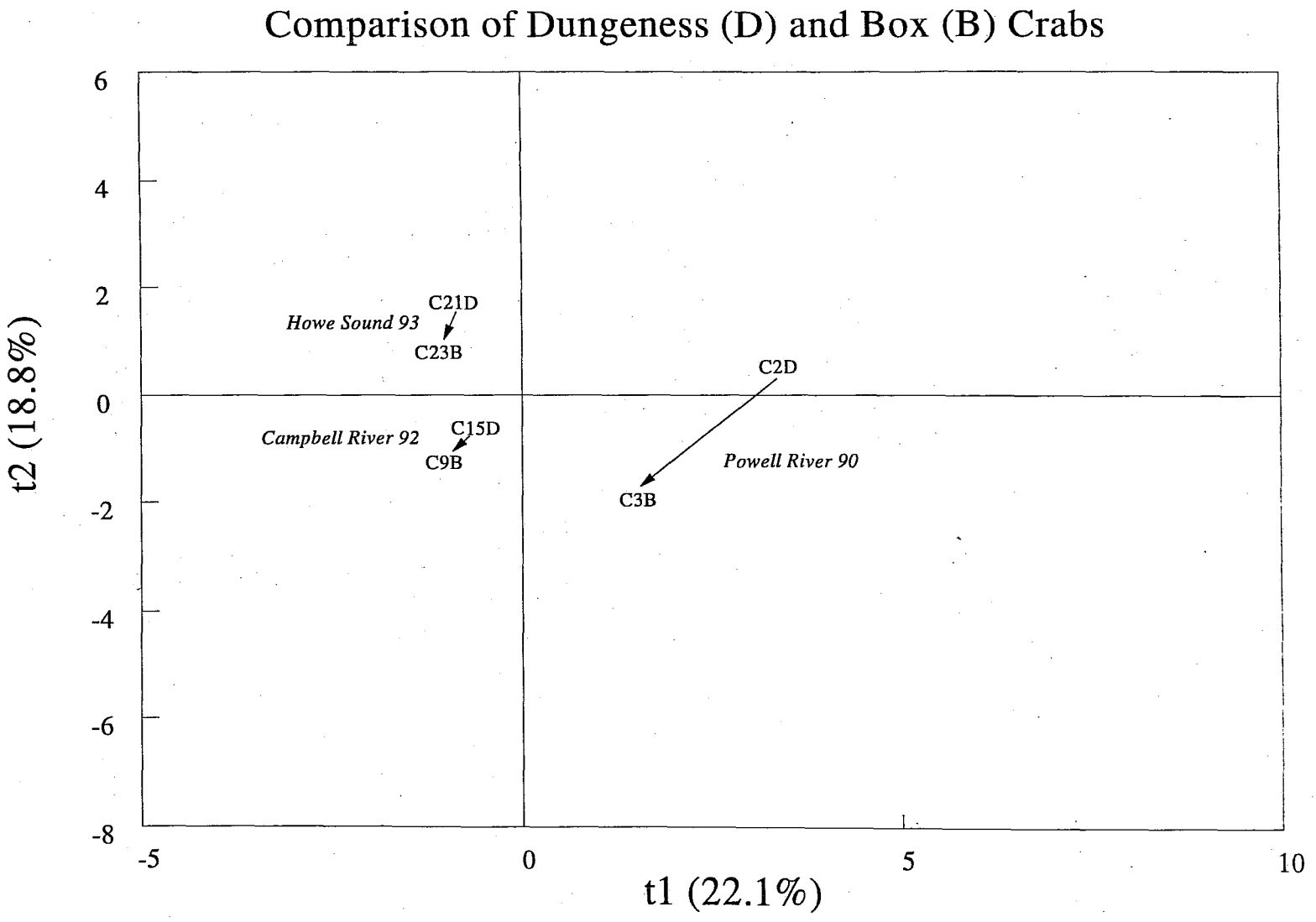

Figure 3-6. Dungeness and Box crab samples from Campbell River, Powell River and Howe Sound.

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4. HARBOURS AND REMOTE AREAS

HARBOURS

Crab samples from Burrard Inlet (Vancouver Harbour), the Fraser River Estuary, Victoria Harbour and Chemanius Harbour were collected by the Environmental Protection Service in July 1987 and analyzed by the Canadian Wildlife Service (Norstrom *et al.* 1988; see Map 1-1 for area locations). The remaining harbour samples were collected by the Department of Fisheries and Oceans or, in a few cases, by the Environmental Protection Service. Samples were obtained from Burrard Inlet in June, October and November 1990 and January 1992, the Fraser River estuary in February 1992 and March 1993 and Victoria and Esquimalt Harbours in March of 1991, 1992 and 1994. Analytical laboratories are detailed in Table 4-1. All harbour samples were Dungeness crab.

Harbour samples from all years and all locations project together in a single cluster (Figure 4-1). This cluster comprises samples from Vancouver Harbour (including Burrard Inlet, False Creek, Port Moody and Indian Arm), the Fraser River estuary, Victoria Harbour (including also Esquimalt Harbour and Esquimalt Lagoon) and Chemainus Harbour (Figure 4-2 to Figure 4-5; Table 4-1). Samples from Nanaimo Harbour and Prince Rupert Harbour also project in or near to the cluster (see Sections 9 and 12). There are no apparent differences in PCA projections between years or between locations in each area — e.g., Indian Arm, Burrard Inlet and False Creek all have similar compositions. Hence crabs from the harbours must have been exposed to PCDD/Fs with a similar composition and/or a similar source; likely candidates include pentachlorophenol wood preservatives and combustion processes.

Two samples collected in 1987 from locations outside of the major harbours have slightly different projections. The sample to the lower right of the cluster (F87) was obtained from Iona Island, near the Fraser River estuary; a subsequent sample from this location in 1992 projected with the other harbour samples. The sample just below the centre of the cluster (Ch87) was obtained from Chemainus Harbour; this sample has a composition that is different from samples obtained from just outside the harbour in the Crofton mill receiving environment (Figure 4-5; Map 6-1 and Section 6).

Composition profiles for the major harbours have not changed dramatically over the five to seven years between the earliest and most recent samples (Figure 4-2 to Figure 4-4). Changes in composition which do occur are primarily for congeners that do not make a strong contribution to projections in the first two PCs; this contributes to the lack of change observed in the harbour PCA projections over time (Figure 4-1). However, congener totals and TEQs have decreased in all of the major harbours, both at individual sampling locations and in the harbours as a whole (Table 4-1). In all three locations the most noticeable change in composition is an increase in the proportions of non-2,3,7,8 substituted congeners for the HpCDDs and the TCDFs, PnCDFs and HxCDFs. Proportions of the other hepta- and octachlorinated congeners have also increased

noticeably in most cases. These congeners are prominent constituents in samples of soil and sludge and in some samples of roof dust from the Alcan aluminum smelter site in Kitimat — a linkage of these congeners to combustion processes is implied (see Section 13 and Figure 13-2). Accordingly, the increase in the proportion of these congeners suggests that combustion byproducts are providing an increasingly important contribution to contaminant profiles for the harbours. Data are not available to distinguish whether the input of these congeners has increased since 1987, or whether they were present all along and are only becoming apparent as concentrations of the non-combustion congeners decrease.

REMOTE AREAS

Baseline crab samples from remote areas of the B.C. coast, including the Queen Charlotte Islands, the Central Coast and Phillips Arm, were collected by the Department of Fisheries and Oceans in March 1991 and analyzed by the Burlington Ontario laboratory (see Map 1-1 for area locations). Samples collected on the Central Coast and in the Queen Charlotte Islands had too few congeners detectable to be retained in the PCA model (Table 2-2). Subsequently Fisheries and Oceans resampled Phillips Arm and collected crab samples from Quatsino Sound in December 1992 and Sidney Island (near Sidney on Vancouver Island) in March 1993. Samples were also obtained from commercial catches in locations near Nootka Sound and the Queen Charlotte Islands in May 1993, Clayoquot Sound in June 1993 and the Prince Rupert area in August 1993 (the latter sample is discussed in Section 12). Analytical laboratories are detailed in Table 4-1. All remote area samples were Dungeness crab.

Baseline Locations

Samples from Clayoquot Sound, Sidney Island and Nootka Sound have similar PCA projections and congener compositions (Figure 4-6 to Figure 4-8). While these samples have been collected in locations away from the mill monitoring sites, the presence of non-2,3,7,8 HxCDDs and of the two TCDFs as the major congeners indicates that these areas may have been impacted by mill discharges. The Nootka Sound sample has a higher congener concentration and TEQ and may have been influenced by discharges from the Gold River mill (Section 8). The sample from Graham Island in the Queen Charlotte Islands has a noticeably higher proportion of potential combustion congeners (e.g. the non-2,3,7,8 substituted congeners for the HpCDDs and the TCDFs, PnCDFs and HxCDFs; see above), but the presence of a substantial HxCDD component may indicate additional input from pentachlorophenol wood preservatives.

It is unlikely that any of these samples represent a background/atmospheric input pattern for crabs on the B.C. coast. One may have to sample even more remote locations (e.g. a seamount) to find an area that has not been impacted by mill effluents or wood preservatives.

To facilitate interpretation of PCA results for the mill sites, samples from baseline locations (QC and SI for the Queen Charlotte and Sidney Islands, respectively) and the three major B.C. harbours (V, B and F for Victoria, Vancouver and the Fraser Estuary, respectively) are shown in a smaller point size on the PCA plots in the sections which follow.

Quatsino Sound and Phillips Arm

Quatsino Sound has a pulp mill at Port Alice on Neroutsos Inlet and a large copper mine which discharges tailings into Rupert Inlet. The pulp mill uses sulphite bleaching with mild chlorination in a process that is unique to British Columbia and Canada (Hatfield Consultants Ltd., 1994). Samples have both congener profiles and PCA projections that are the same as the major harbours (Figure 4-1, Figure 4-6 and Figure 4-9). Congener concentrations and TEQs are low and comparable to other remote locations around Vancouver Island. Hatfield Consultants Ltd. (1994) have attributed the low levels of dioxins and furans in the mill effluent to a lack of dioxin precursors and the combination of a relatively mild chlorination treatment and the low lignin content unbleached sulphite pulps. As with the Gold River samples (Section 3), the congener pattern also indicates either that very few pentachlorophenol contaminated chips were used or that pulping conditions did not favour HxCDD formation.

Samples from Phillips Arm have a different PCA projection and congener profile than other remote areas (Figure 4-6 and Figure 4-10). Congener concentrations and TEQs are also much higher than expected for a remote area. This location is presently the site of a logging camp and has apparently been contaminated by chemicals with an unknown source.

Table 4-1. Samples from B.C. harbours and remote areas of the B.C. coast.

Samples Averaged	Laboratory [a]	Congener Total	Calculated TEQ
		pg/g wet weight	
Harbours — Earliest Samples			
Burrard Inlet, 1987 (n=1)	CWS	650	27
Fraser River Estuary, 1987 (n=1)	CWS	860	73
Victoria Harbour, 1987 (n=1)	CWS	3600	130
Chemanius Harbour, 1987 (n=1)	CWS	1200	69
Harbours — Most Recent Samples			
Burrard Inlet, 1992 (n=7)	BUR	380	14
Fraser River Estuary, 1993 (n=4)	AXYS	430	16
Victoria Harbour, 1994 (n=6)	IOS	1500	38
Remote Areas			
Graham Is., Clayoquot Sound., Sidney Is., 1993 (n=4)	IOS, AXYS	21	1.2
Nootka Sound, 1993 (n=1)	IOS	82	5.4
Quatsino Sound, 1992 (n=5)	IOS	33	1.1
Phillips Arm, 1991 and 1992 (n=4)	BUR, IOS	480	24

[a] Laboratories are the Department of Fisheries and Oceans at the Institute of Ocean Sciences in Sidney B.C. (IOS), Axys Analytical Ltd. (AXYS), the Great Lakes Laboratory for Fisheries and Aquatic Sciences in Burlington Ontario (BUR) and the Canadian Wildlife Service (CWS).

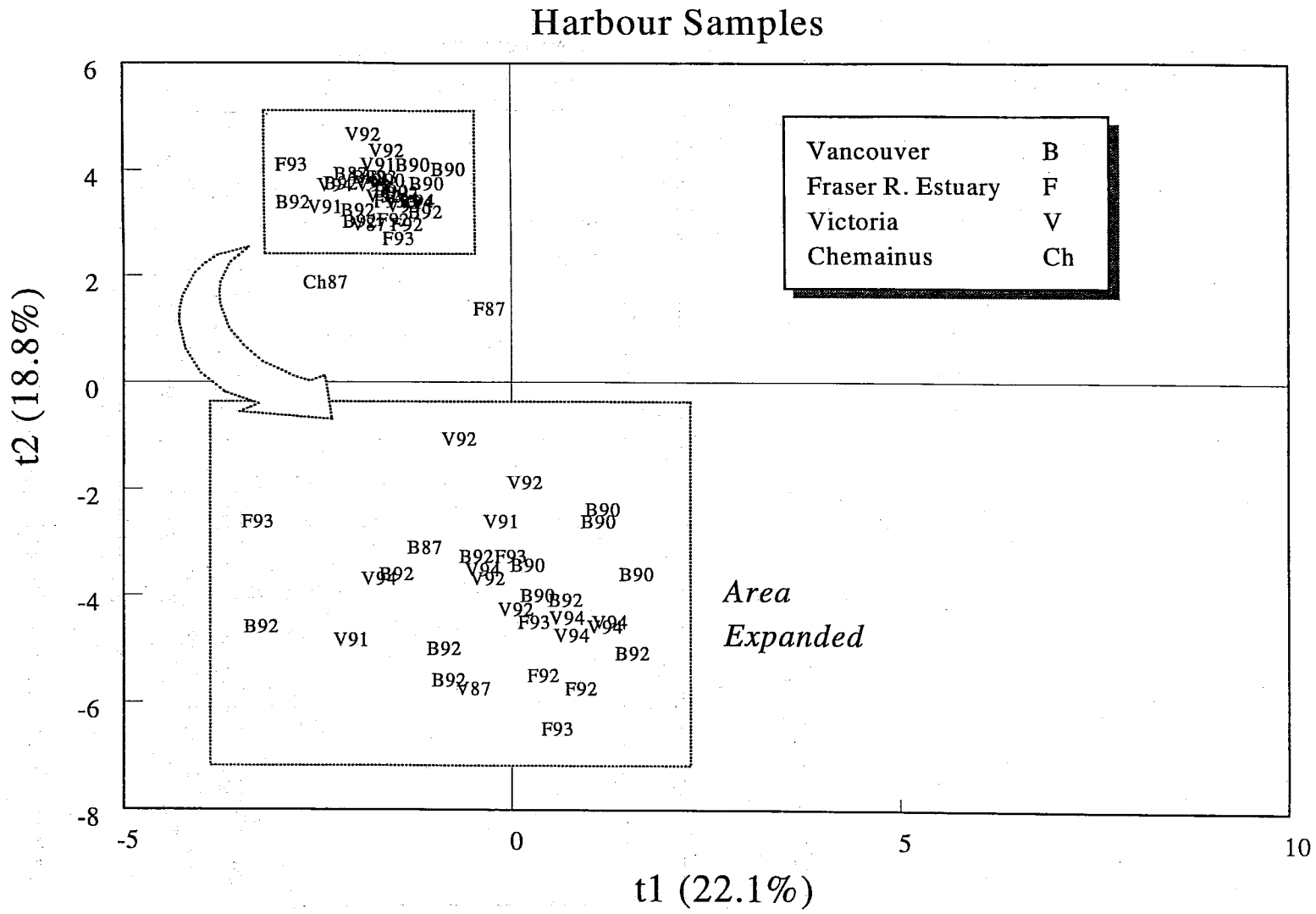


Figure 4-1. PCA projections for samples from the B. C. harbours.

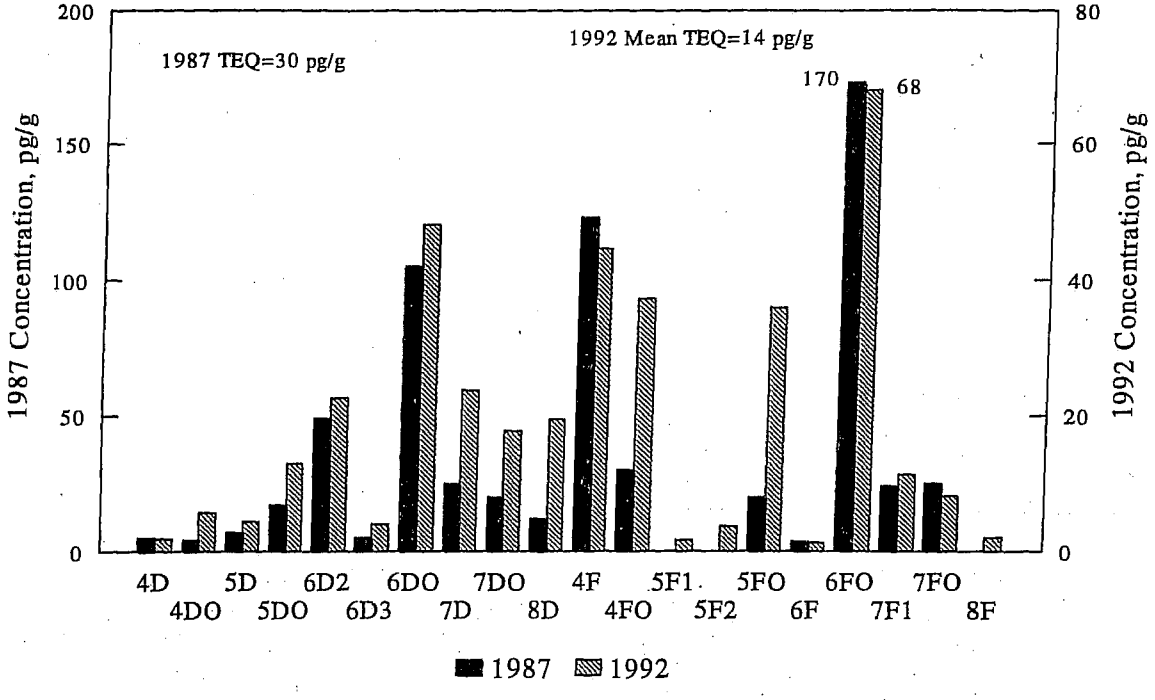


Figure 4-2. Vancouver Harbour/Burrard Inlet in 1987 (n=1) and 1992 (n=7).

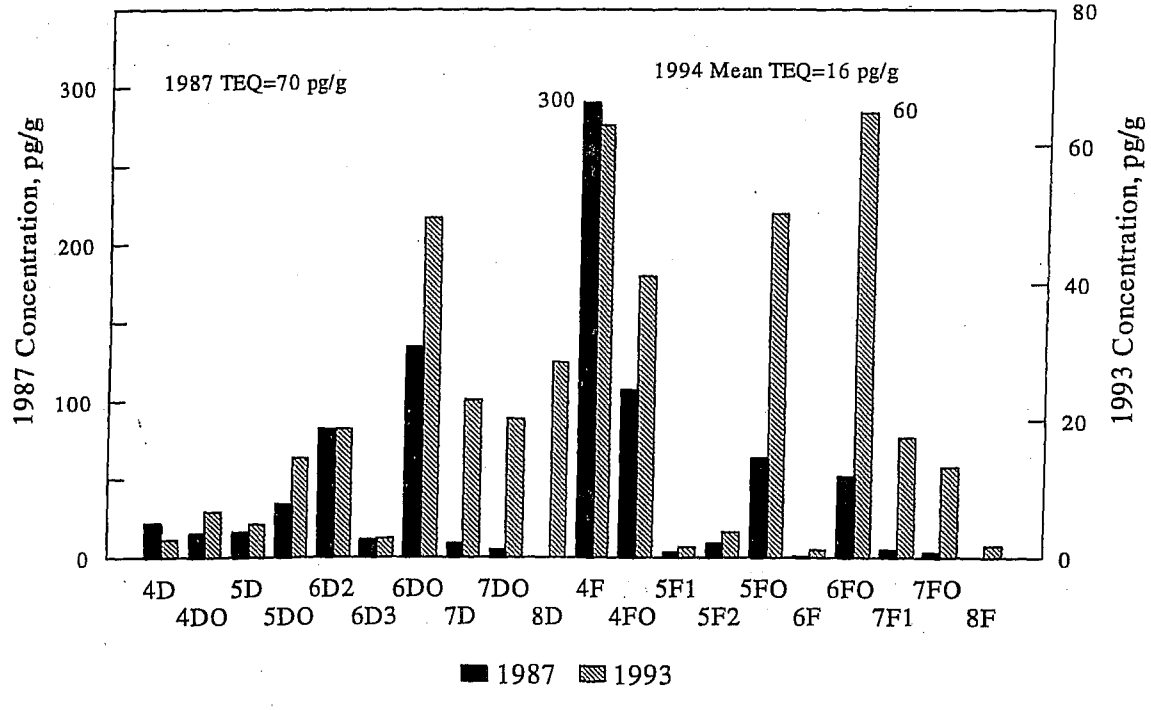


Figure 4-3. The Fraser River estuary in 1987 (n=1) and 1993 (n=4).

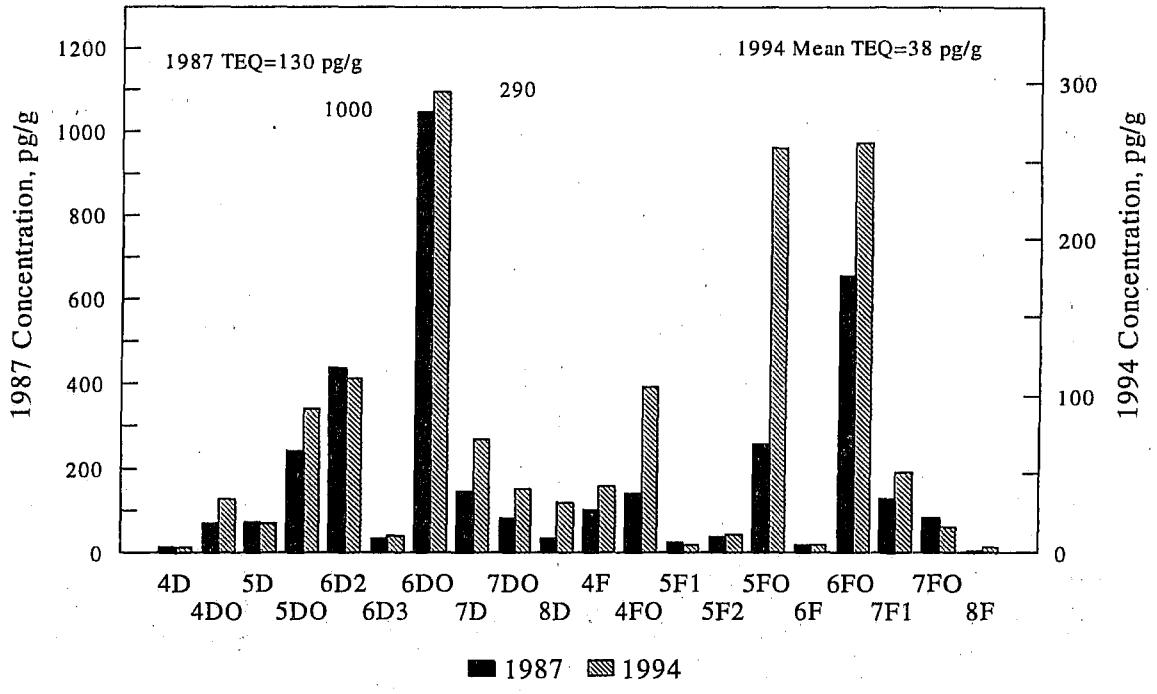


Figure 4-4. Victoria and Esquimalt harbours in 1987 (n=1) and 1994 (n=6).

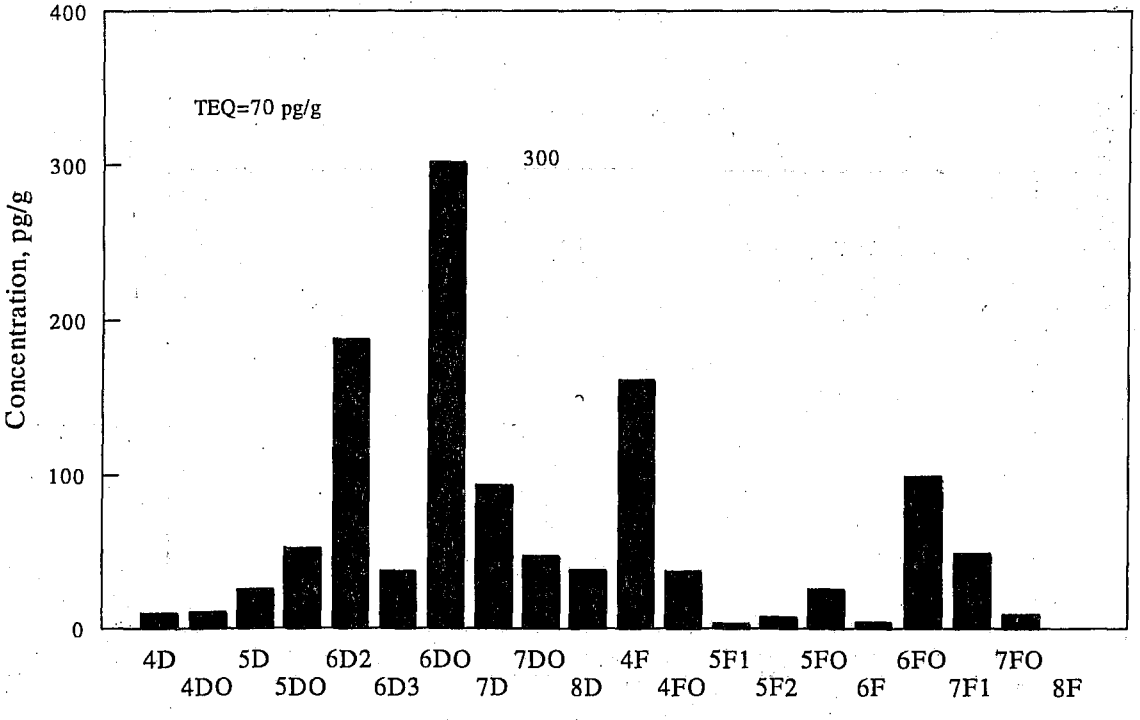


Figure 4-5. Chemanius harbour in 1987 (n=1).

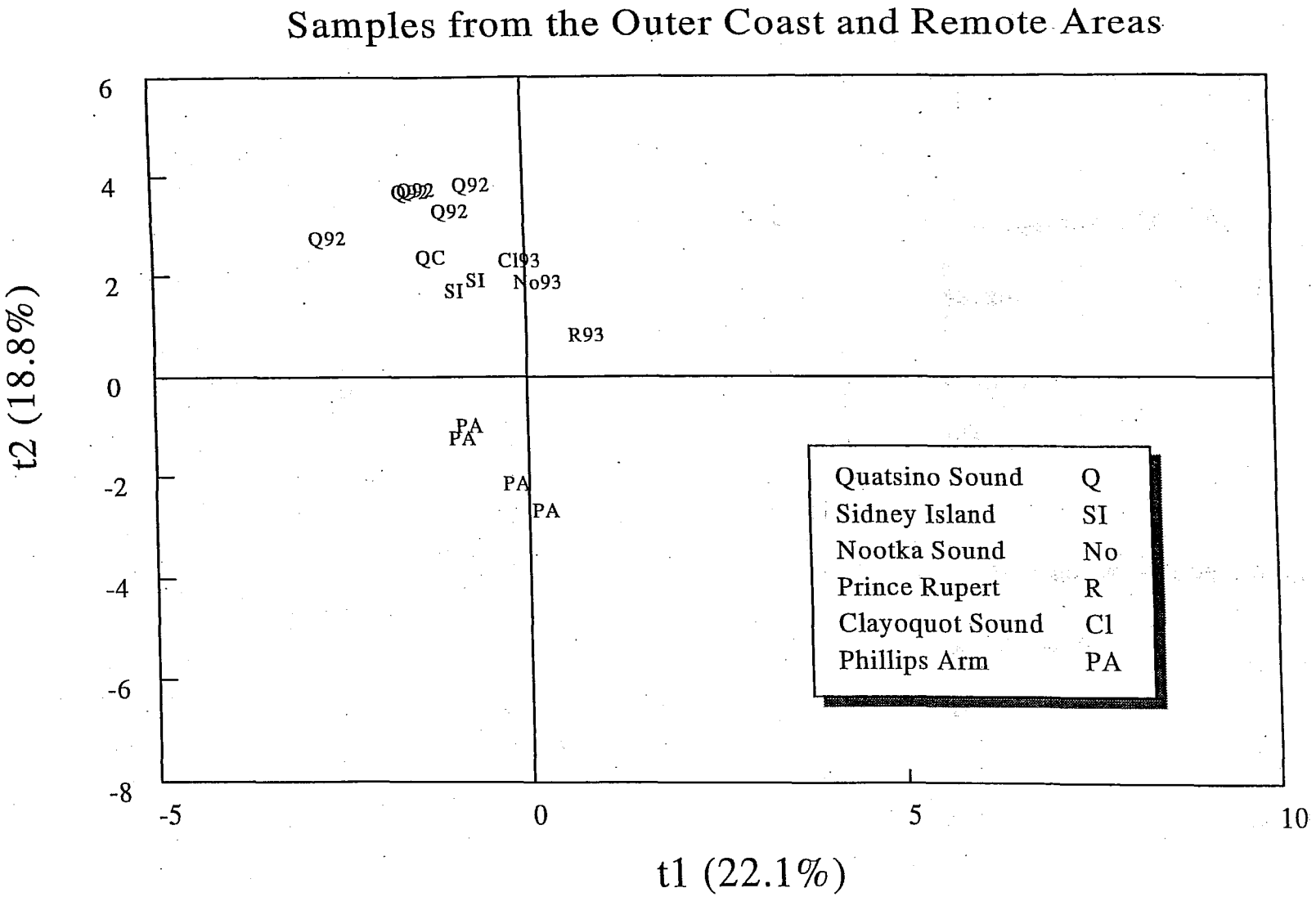


Figure 4-6. PCA projections for samples from remote areas.

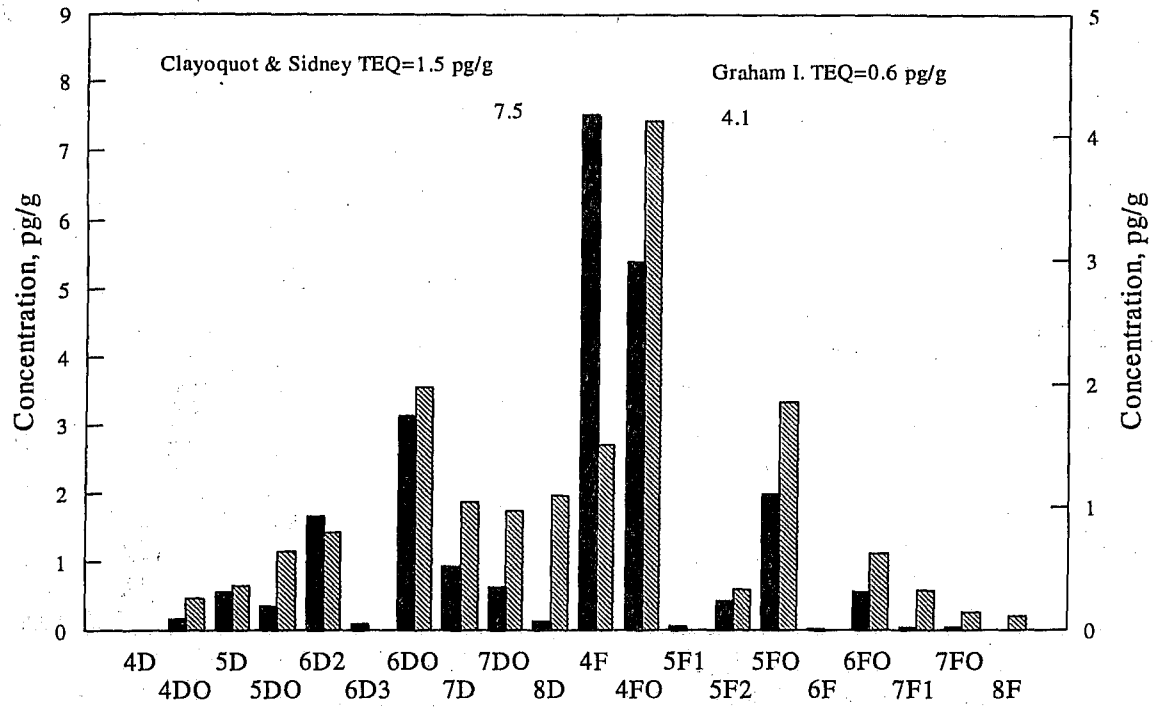


Figure 4-7. Samples collected from remote areas of the B. C. coast in 1993.

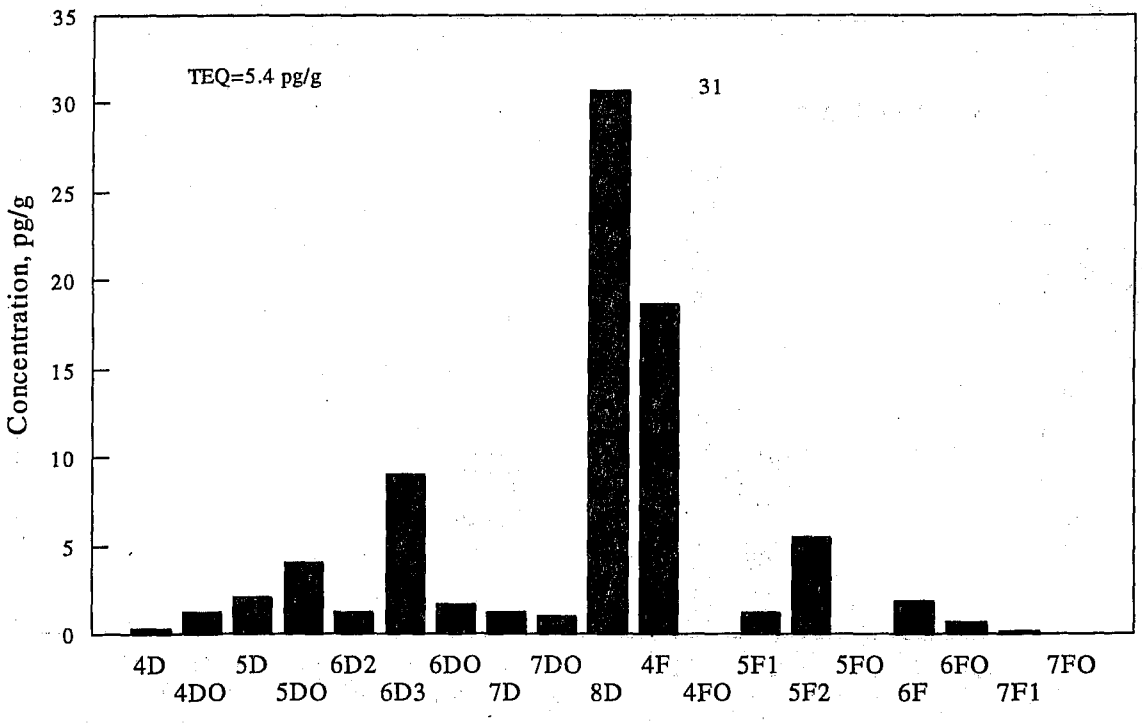


Figure 4-8. Congener profile for a crab sample from Nootka Sound in 1993 (n=1).

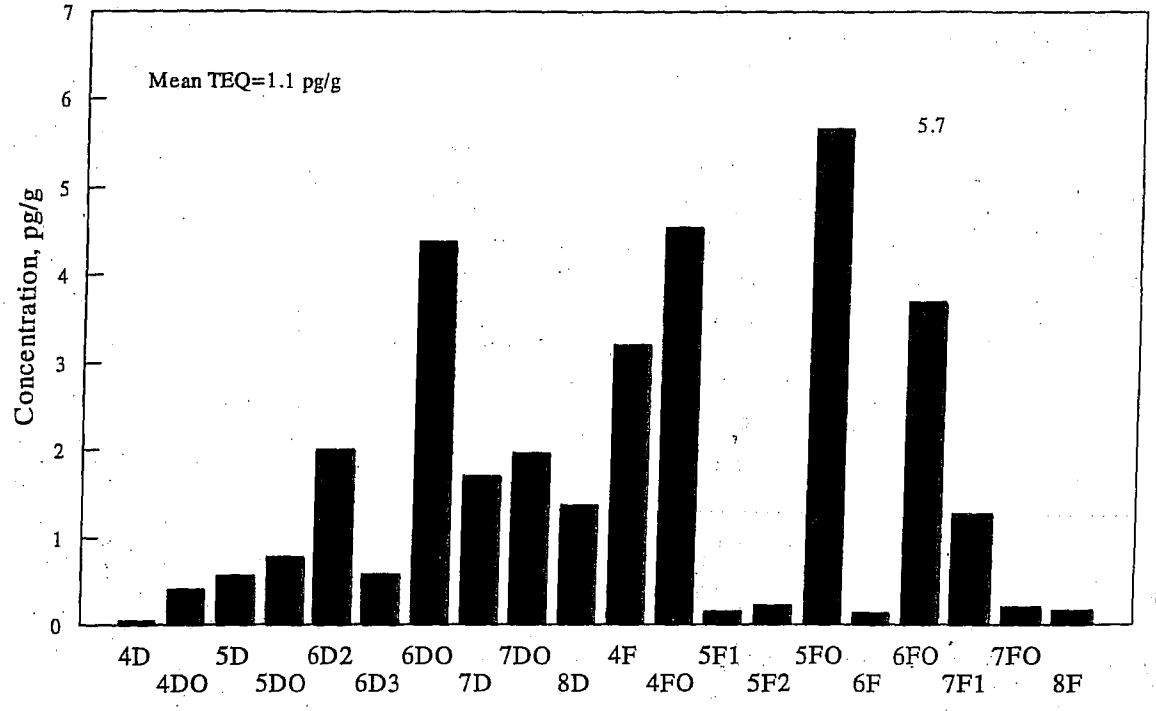


Figure 4-9. Samples from Quatsino Sound (n=5).

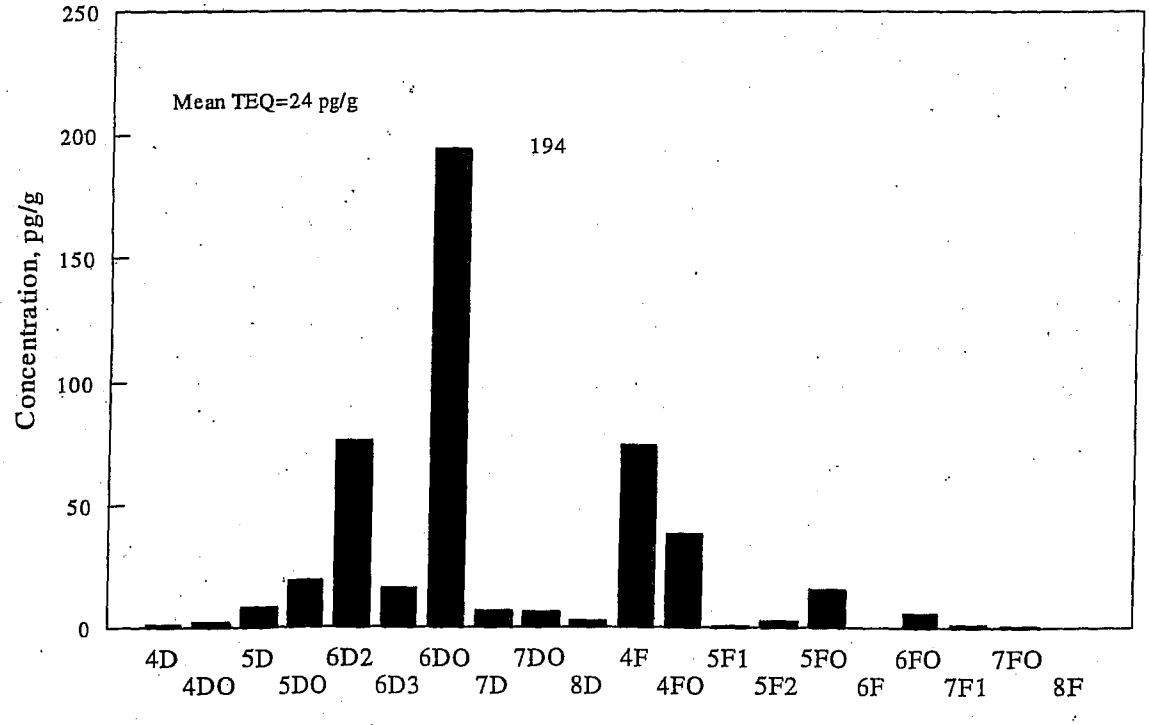


Figure 4-10. Samples from Phillips Arm (n=4).

5. HOWE SOUND

Two pulp mills are located on Howe Sound: the Howe Sound Pulp and Paper Limited mill in Port Mellon and the Western Pulp Limited Partnership mill in Woodfibre, near Squamish (Map 5-1). These mills will be referred to as the Port Mellon and Woodfibre mills, respectively.

Crab samples were collected near the Port Mellon and Woodfibre mills by the Department of Fisheries and Oceans in the winter of 1987-1988 and analyzed by the Canadian Wildlife Service (Norstrom *et al.* 1988). Subsequent samples were collected by Hatfield Consultants Ltd. in both late January/early February and November of 1989, in September 1990, and then annually in February or March from 1991 to 1993 (Map 5-1). These mill monitoring program samples were all analyzed by Axys Analytical Ltd. In March 1993 additional samples were collected by G3 Consulting Ltd. between the Howe Sound and Powell River mill sampling areas and analyzed by the Institute of Ocean Sciences (Map 5-2). All samples from Howe Sound were Dungeness crab except for Box crab from Station C23 in 1993. Samples from outside Howe Sound in 1993 were Dungeness crab from Stations C301 and C303 but were Red Rock crabs from Stations C302, C304, C306 and C309.

Congener profiles and PCA projections for the earliest sampling times indicate only a small change in congener composition between 1987 and January/February 1989 (Figure 5-1 and Figure 5-3). Samples collected close to the Port Mellon mill (P87, C10, C11 and C12) had essentially the same PCA projections in both years while Woodfibre samples (W87, C3, C4 and C5) showed only a small difference in projection (Figure 5-3). The change between years was principally due to a modest decrease in the proportion of 2,3,7,8-TCDF with a small increase in the non-2,3,7,8 TCDFs and the HxCDDs.

The change in congener profile apparently accelerated between January/February and November 1989; this is reflected in a much larger shift to the upper left in individual PCA projections during this time (Figure 5-3). While this second change apparently represents a larger decrease in the proportion of TCDFs than had occurred between 1987 and early 1989, part of the change could be due to differences in season. A progressive shift to the upper left in PCA projections, indicating a progressive increase in the HxCDD proportion, occurs for samples collected between 1989 and 1993 (Figure 5-3 to Figure 5-7). By 1993 the HxCDD proportion was roughly one-half of the size of 2,3,7,8-TCDF (Figure 5-2). The change in composition over time must have proceeded uniformly throughout Howe Sound because PCA projections indicate that the congener composition has remained relatively consistent at any given time (Figure 5-3 to Figure 5-7). The uniformity in composition extended to samples collected well outside Howe Sound in 1993 (see following).

Over the six years of sampling in Howe Sound, the relatively small change in congener profile has been accompanied by a large decrease in both concentration and TEQ (Table 5-1; Figure 5-2). Despite the relatively uniform congener composition that has persisted throughout Howe Sound in each year, samples collected from locations adjacent the Woodfibre mill have

maintained a consistently lower congener total and TEQ than samples from locations adjacent to the Port Mellon mill (Map 5-1).

SAMPLES FROM OUTSIDE HOWE SOUND

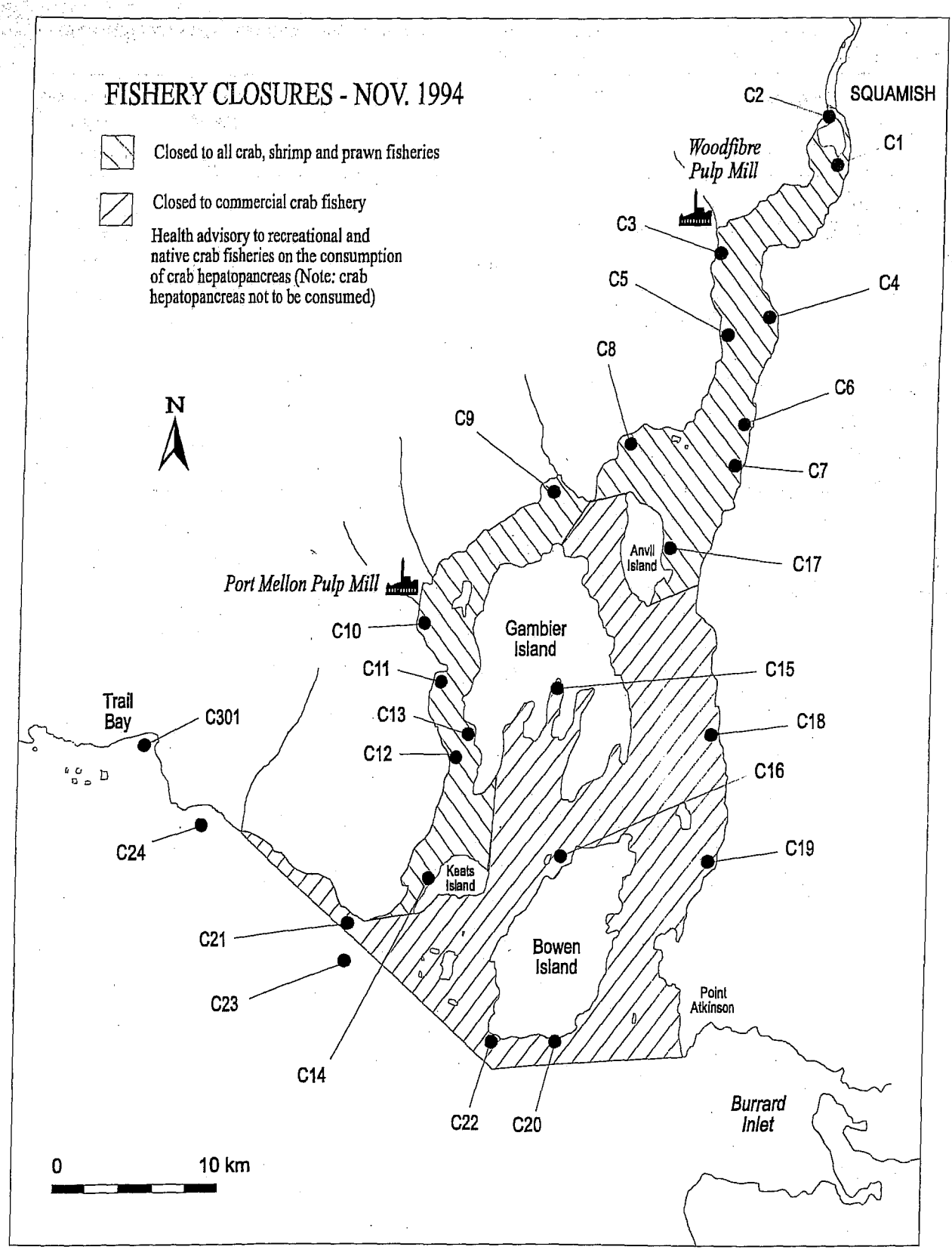
The 1993 PCA projections for Dungeness crab samples from Stations C21 (two samples) and C23 at the mouth of Howe Sound are identical to the PCA projections for Station C301 (Trail Bay; two samples) and nearby Station C24 (Roberts Creek) located to the west of Howe Sound (Figure 5-7; Map 5-2). At a further distance from Howe Sound, the Dungeness crab sample from C303 (Secret Cove) has a composition closer to that of the principal B.C. harbours, which suggests some contribution from a non-mill source.

The Red Rock crab from Station C309 (Halibut Bank) projects further to the right and much closer to the Dungeness crab samples from within Howe Sound (Figure 5-7). While the change in species may contribute to a shift in congener profile to a higher proportion of the most toxic congeners (i.e. a shift to the right in PCA projection; see Section 3), the PCDD/Fs in the C309 crab sample likely have primarily a mill source, and geography would suggest that the Howe Sound mills are the most probable candidates (Map 5-2). In support, positions of the Dungeness crab samples C301 and C303 and the Red Rock crab sample C309 are unchanged relative to the Howe Sound samples in the third PC (not shown). This congruence in three PCs strengthens the assertion that these samples have the same PCDD/F sources as the Howe Sound samples.

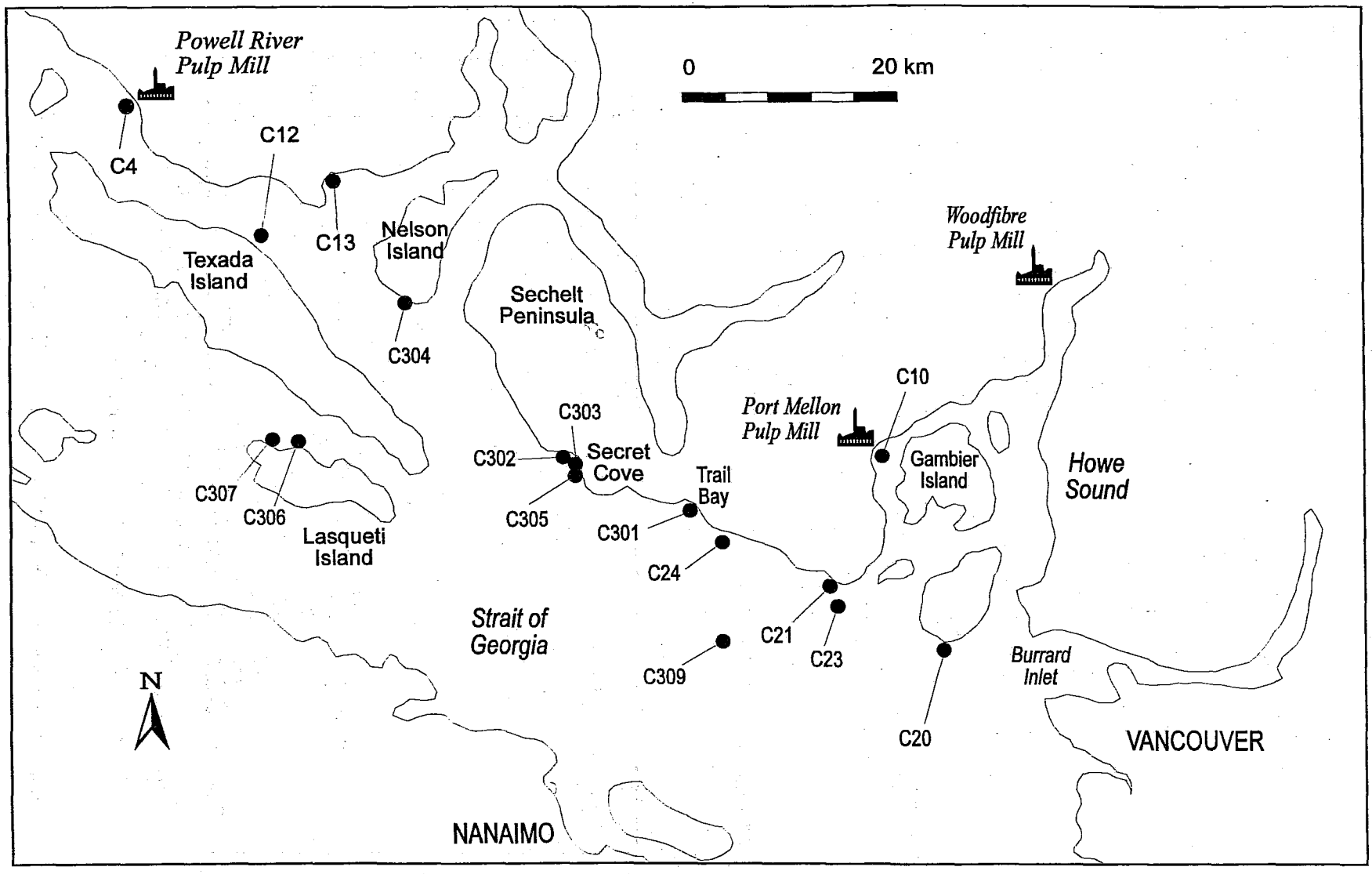
The Red Rock crab sample from Station C302 (Secret Cove), and the samples from Stations C304 and C306 (Nelson Island and Lasqueti Island respectively) project near to the Dungeness crab sample C303 (Secret Cove; Figure 5-7; Map 5-2). The 1993 Dungeness crab sample from Powell River that is closest to Howe Sound also projects with this group of samples (Powell River C13; Map 5-2 and Section 11). However, all of these samples are shifted relative to both the Howe Sound and Powell River samples in the third PC. Projections of these samples in the third PC indicate a higher proportion of the hepta- and octachlorinated congeners than other samples from the Howe Sound region. This shift could be due to either a decrease in mill influence in the region between the two mills and/or an additional source of PCDD/Fs.

Table 5-1. Dungeness crab hepatopancreas congener total and TEQ averages for samples with similar PCA projections from Howe Sound.

Samples Averaged	**Congener Total**	**Calculated TEQ**
	pg/g wet weight	
Earliest Samples		
Port Mellon and Woodfibre mills, 1987 (n=2)	30,000	2900
Howe Sound C3, C4, C5, C10, C11 and C12, Jan/Feb 1989 (n=6)	9900	850
Howe Sound, 1987 (n=2) plus C3, C4, C5, C10, C11 and C12, 1989 (n=6)	15,000	1400
Most Recent Samples		
Port Mellon C10 and C14, 1993 (n=6)	1300	120
Woodfibre C3 and C5, 1993 (n=2)	79	7.0
Port Mellon C10 and C14 (n=6) plus Woodfibre C3 and C5, 1993 (n=2)	1000	95



Map 5-1. Crab sampling locations in Howe Sound.



Map 5-2. Crab sampling locations between Howe Sound and Powell River.

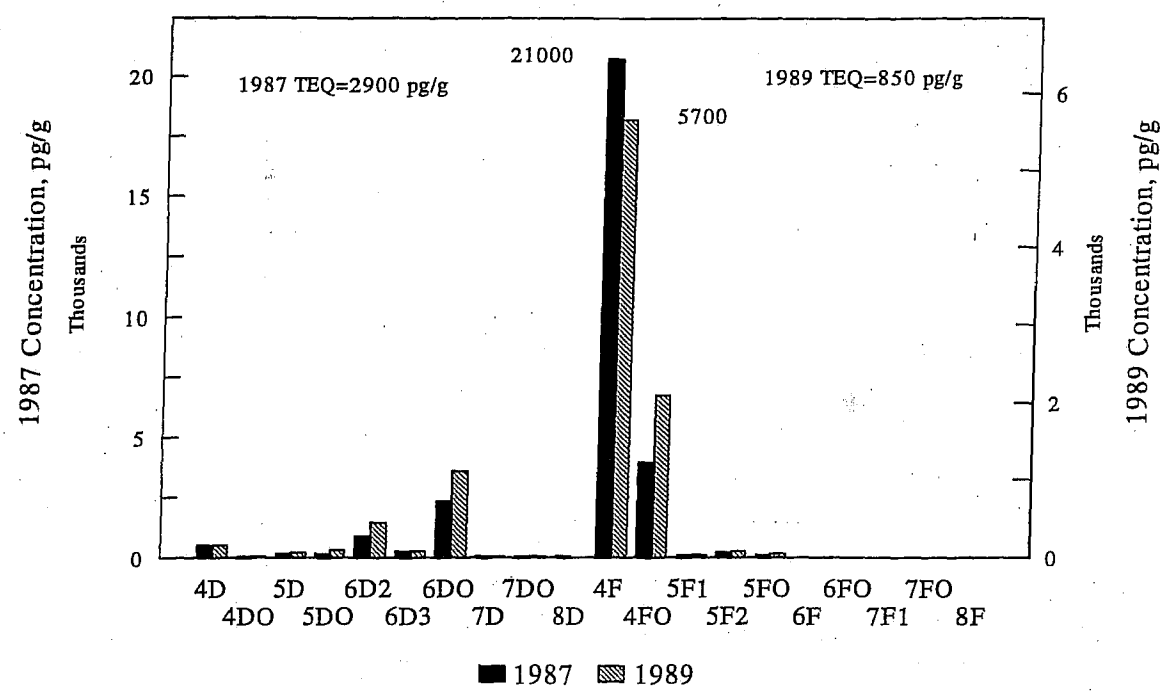


Figure 5-1. Howe Sound congener profiles for 1987 (n=2) and 1989 (n=6).

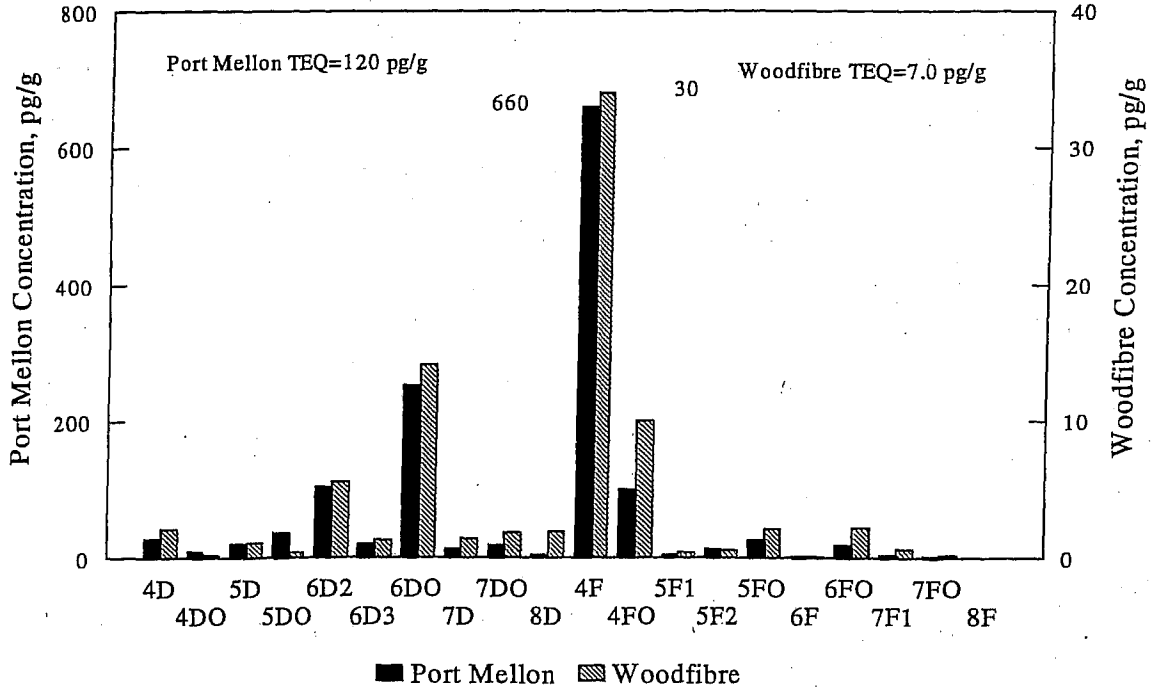


Figure 5-2. Congener profiles for Port Mellon (n=6) and Woodfibre (n=2) mills in 1993.

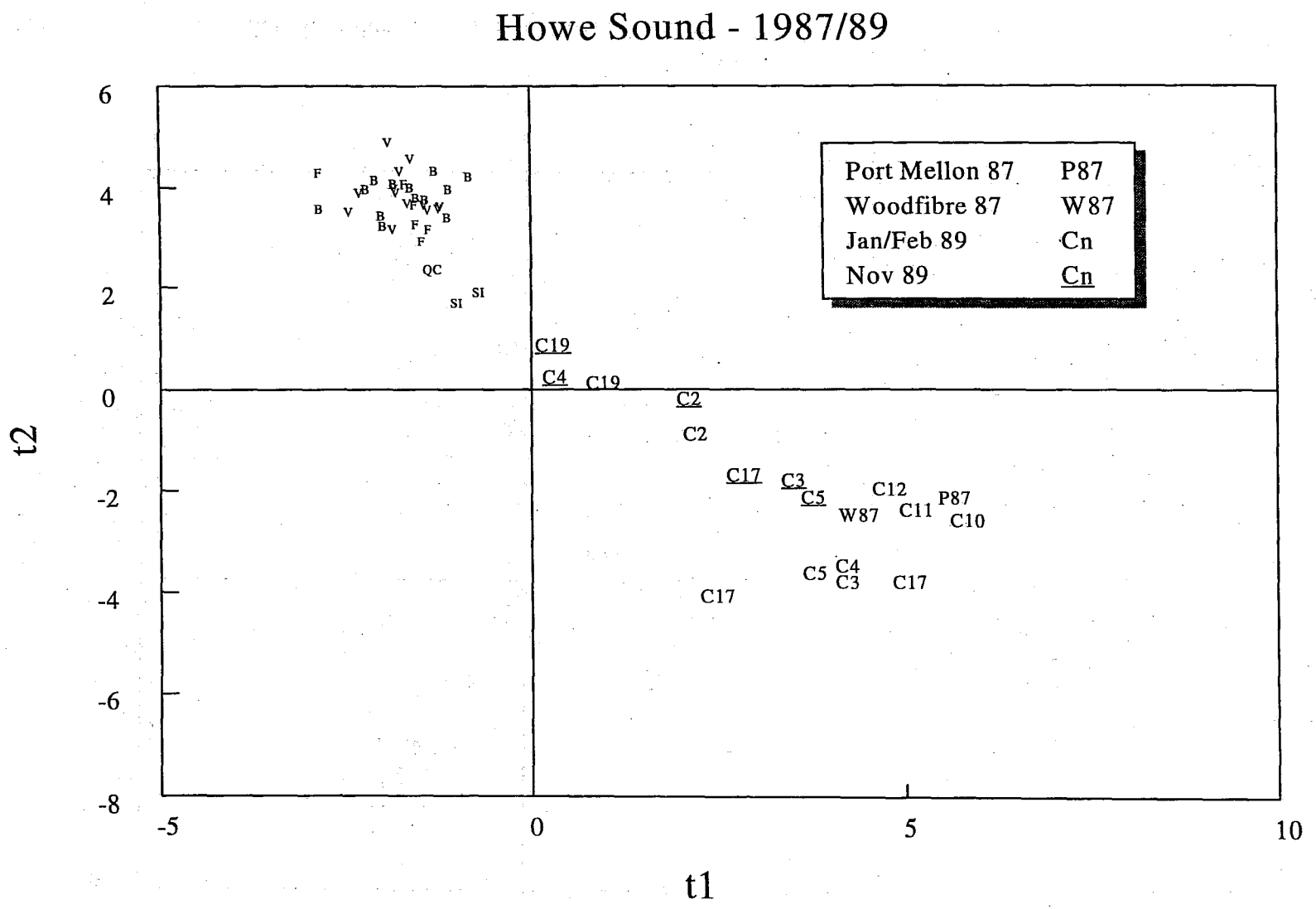


Figure 5-3. PCA projections for samples from Howe Sound in 1987 and 1989.

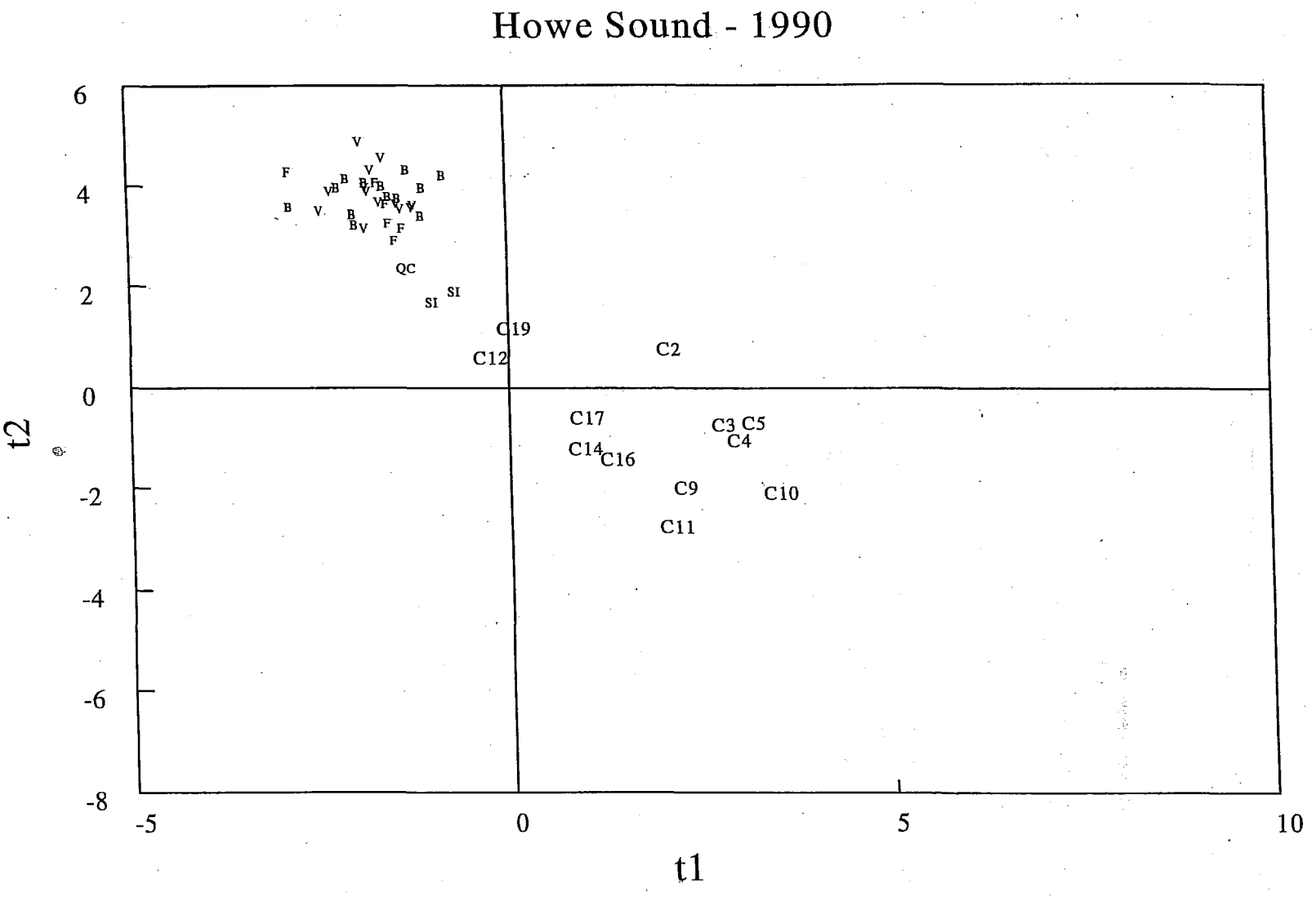


Figure 5-4. PCA projections for samples from Howe Sound in 1990.

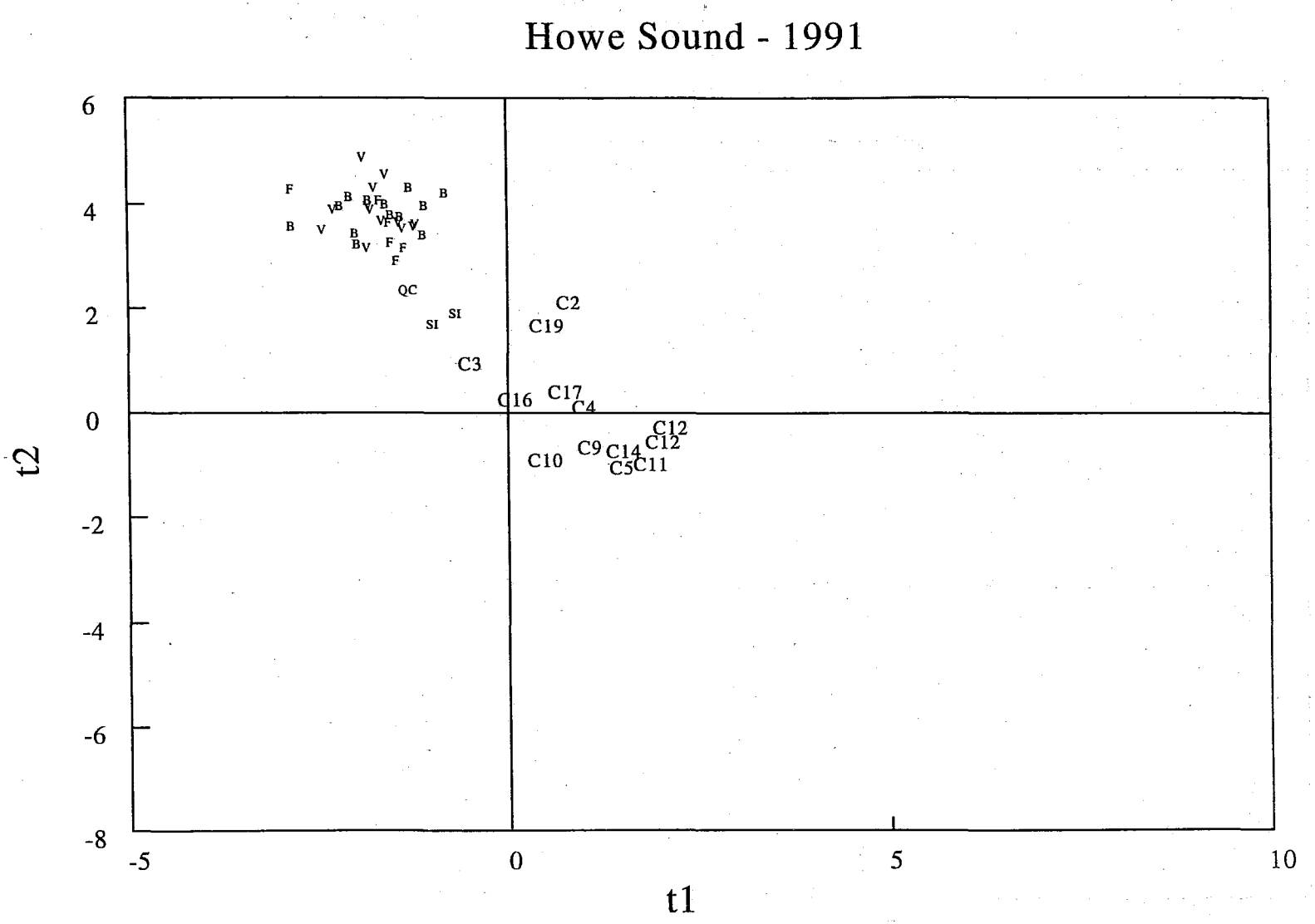


Figure 5-5. PCA projections for samples from Howe Sound in 1991.

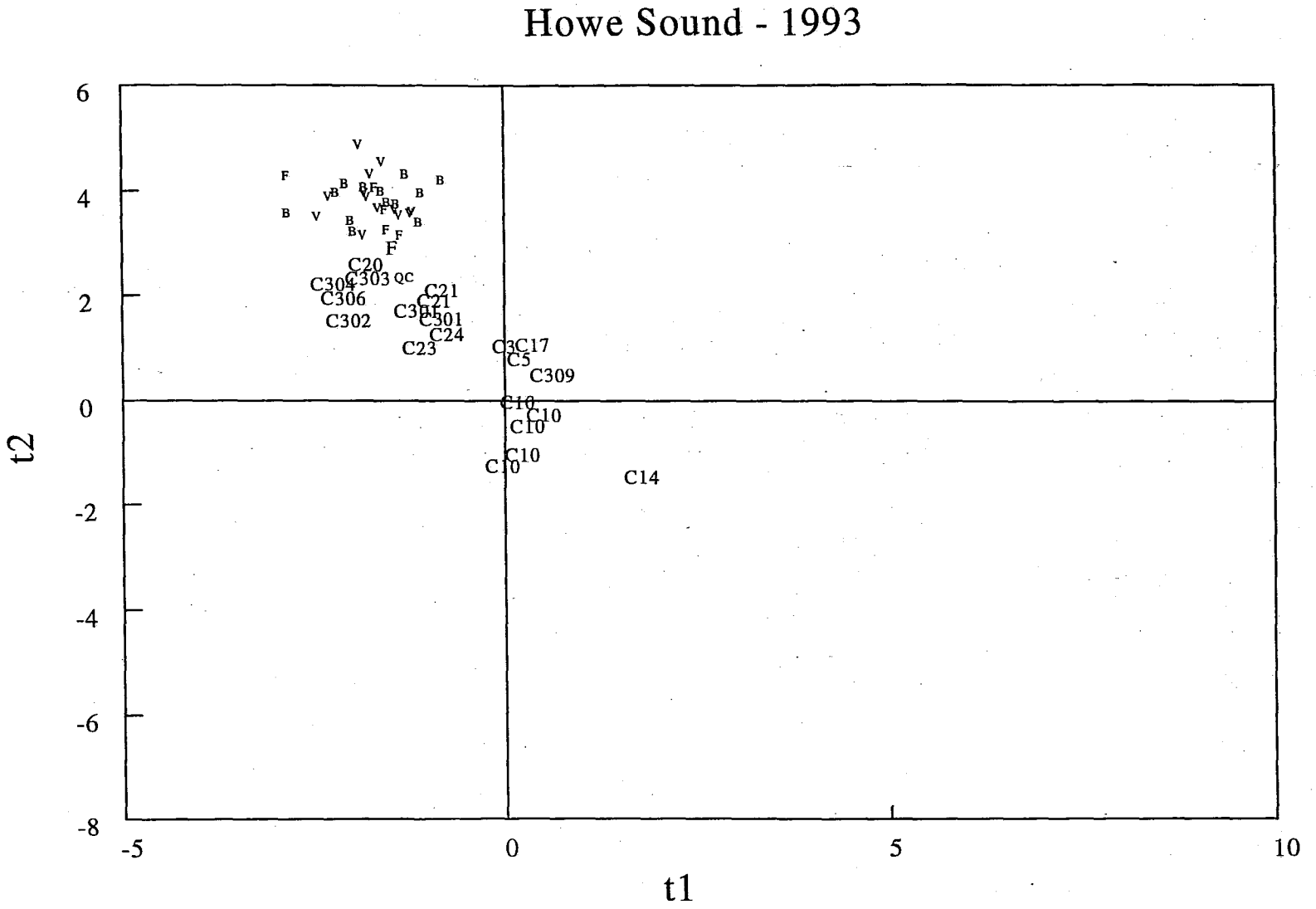


Figure 5-7. PCA projections for samples from Howe Sound in 1993.

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6. CROFTON

Crab samples adjacent to the Fletcher Challenge Canada Limited Crofton Pulp and Paper mill were collected by the Environmental Protection Service in July 1987 and analyzed by the Canadian Wildlife Service (Norstrom *et al.* 1988). The next samples were collected north and south of the Crofton mill by Hatfield Consultants Ltd. in January 1990 and at the head of Cowichan Bay by Envirochem Ltd. in January and March 1990 (Map 6-1). The sampling range was extended further to the north above Thetis Island and to the south into Saanich Inlet by Hatfield Consultants Ltd. in August 1990; six of these samples (C14, C15, C16, C16A, C19 and C21) had too few congeners detectable to be retained in the PCA model (Table 2-2). Subsequent Crofton mill monitoring samples were collected by Hatfield Consultants Ltd. in April 1991, February 1992 and March 1993. Axys Analytical Ltd. analyzed all mill monitoring program samples from 1990 to 1993 except for the two 1990 Envirochem samples from Cowichan Bay that were analyzed by B.C. Research. All samples from the Crofton vicinity have been Dungeness crab.

Between 1987 and 1990 the calculated TEQ decreased markedly for stations closest to the mill (Table 6-1). Congener profiles and PCA projections both indicated a decrease in the proportion of toxic congeners over this time (Figure 6-1 and Figure 6-4). The change in composition between years was principally due to a decrease in the TCDFs coupled with an increase in the HxCDDs. PCA results indicate that PCDD/Fs with a pulp mill origin were present in 1990 as far north as Ladysmith and as far south as Saanich Inlet (Figure 6-4).

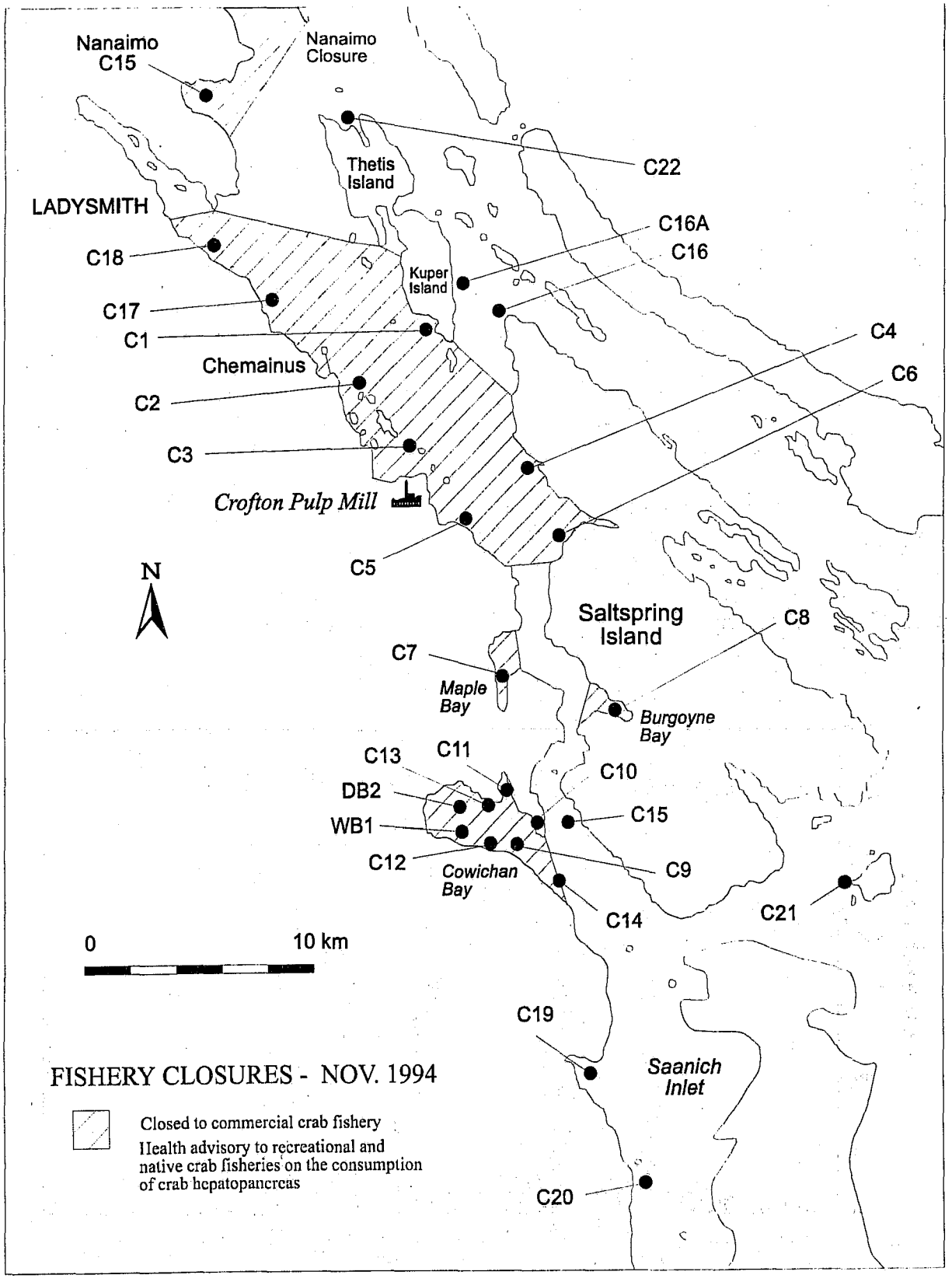
From 1990 through to 1993 samples close to the Crofton mill exhibited only a small decrease in the congener total and calculated TEQ (Table 6-1). PCA projections and congener profiles also showed only a small change from year to year, with the major change being a decrease in the proportion of the TCDFs (Figure 6-3; Figure 6-5 to Figure 6-7). In 1993 samples from Thetis Island (C22) in the north, to the Crofton mill (C3 and C4), to Cowichan Bay (C13) in the south had the same congener composition (Figure 6-7), indicating that PCDD/Fs from the Crofton mill have persisted in the region.

Sources of PCDD/Fs other than the Crofton mill become potentially more important at the edges of the Crofton monitoring area (Map 6-1). In the north, PCA projections of crabs from Crofton Stations C22 and C18 and Nanaimo/Harmac Station C15 (see Section 9) overlap — projections are consistent with either mill as a source or with mixed inputs. To the south, samples C12, C13, DB2 and WB1 from within Cowichan Bay in 1990 exhibited congener profiles and PCA projections that were distinct from the Crofton and Saanich Inlet samples (Figure 6-2 and Figure 6-4). The presence of non-2,3,7,8-substituted furans in the Cowichan Bay samples (Figure 6-2) is consistent with the input of pentachlorophenol wood preservatives into Cowichan Bay as a result of sawmill and lumber transshipment activities. In subsequent years samples from Station C10 at the mouth of Cowichan Bay always had a Crofton mill pattern, while Stations C13 and C14 from further back in the bay were similar to the pulp mill in some years, and Cowichan Bay

in others. The results suggest that Crofton mill effluent has made only a partial contribution to the congener profiles exhibited by crabs from Cowichan Bay.

Table 6-1. Dungeness crab hepatopancreas congener total and TEQ averages for samples with similar PCA projections from Crofton.

Samples Averaged	**Congener Total**	**Calculated TEQ**
	pg/g wet weight	
Earliest Samples		
Crofton, 1987 (n=2)	4300	420
Crofton C4, C5, C6, C7 and C8, 1990 (n=5)	1700	130
Crofton 1987 (n=2) plus C4, C5, C6, C7 and C8, 1990 (n=5)	2400	220
Cowichan Bay C12, C13, DB2 and WB1, 1990 (n=4)	470	27
Most Recent Samples		
Crofton C3, C4 and C7, 1993 (n=3)	1500	94



Map 6-1. Crab sampling locations near Crofton.

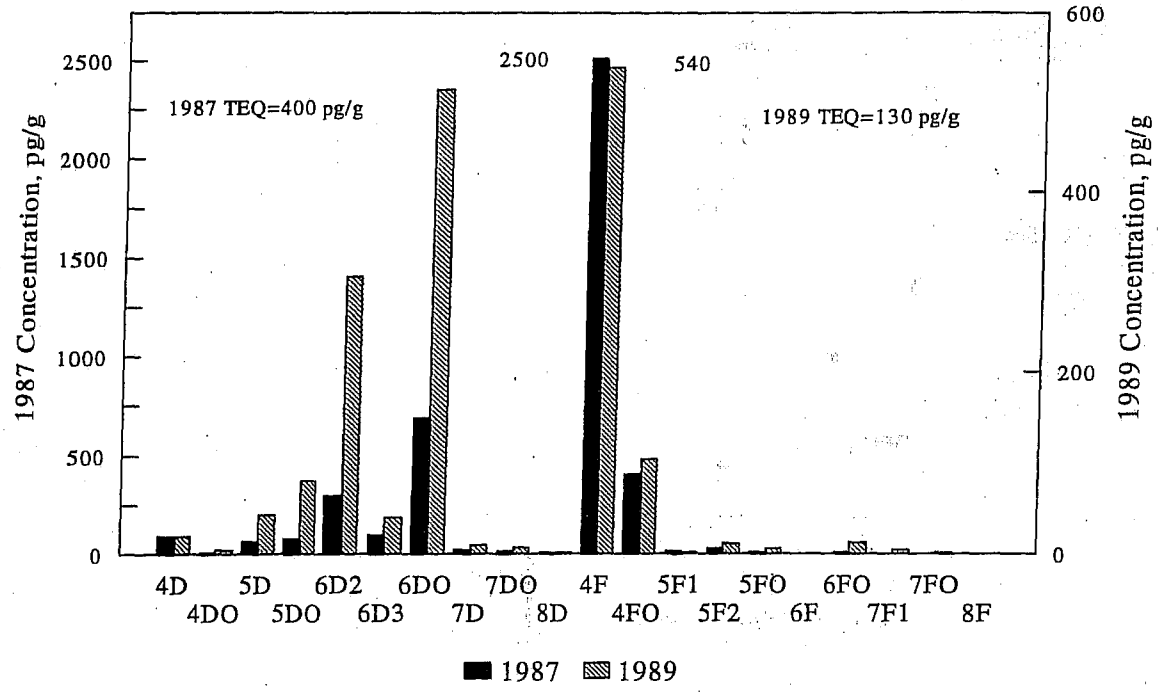


Figure 6-1. Crofton congener profiles for 1987 (n=2) and 1990 (n=5).

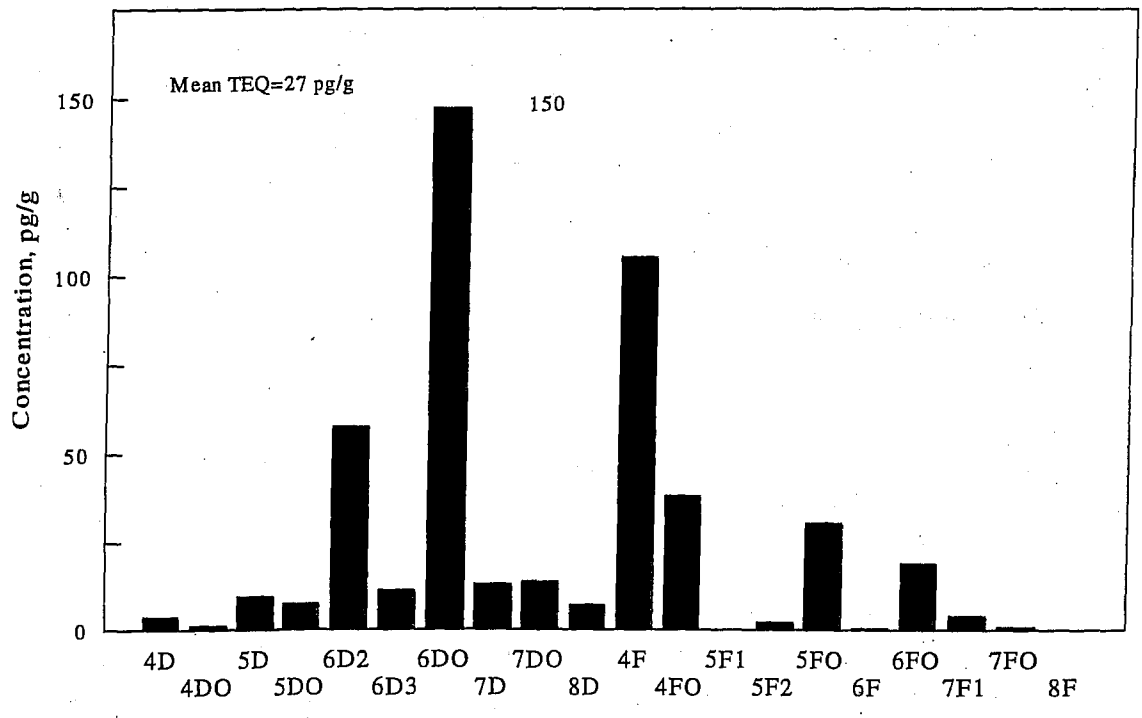


Figure 6-2. Cowichan Bay congener profiles for 1990 (n=4).

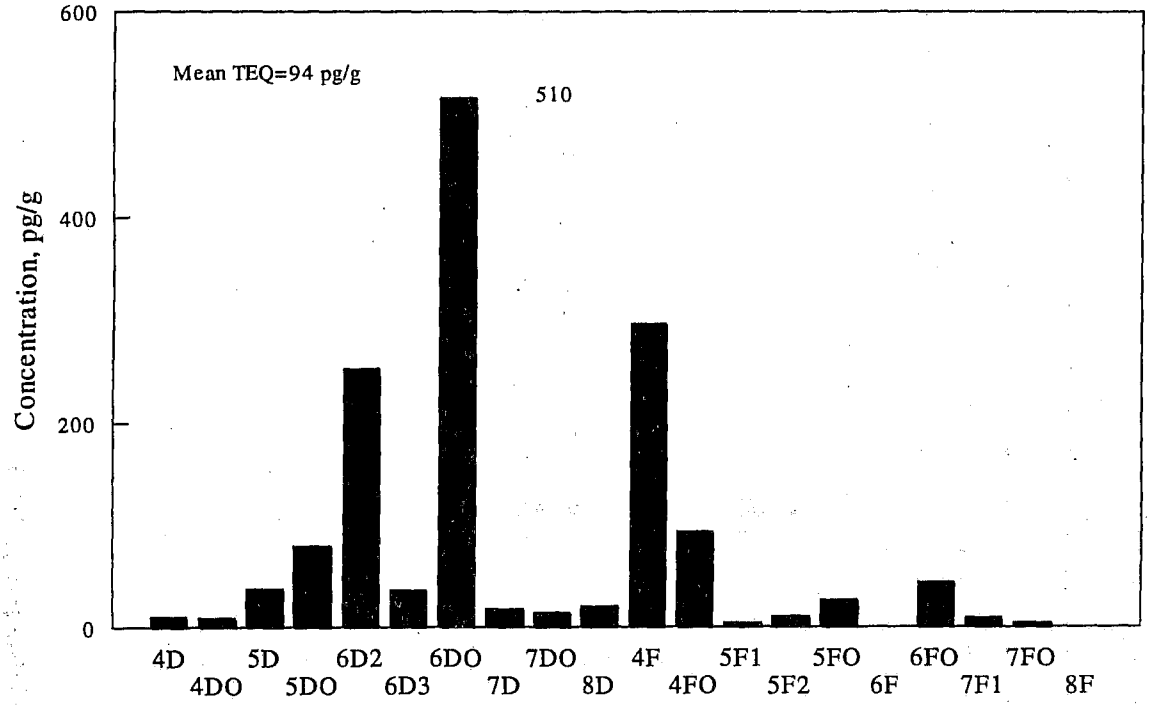


Figure 6-3. Crofton congener profiles for 1993 (n=3).

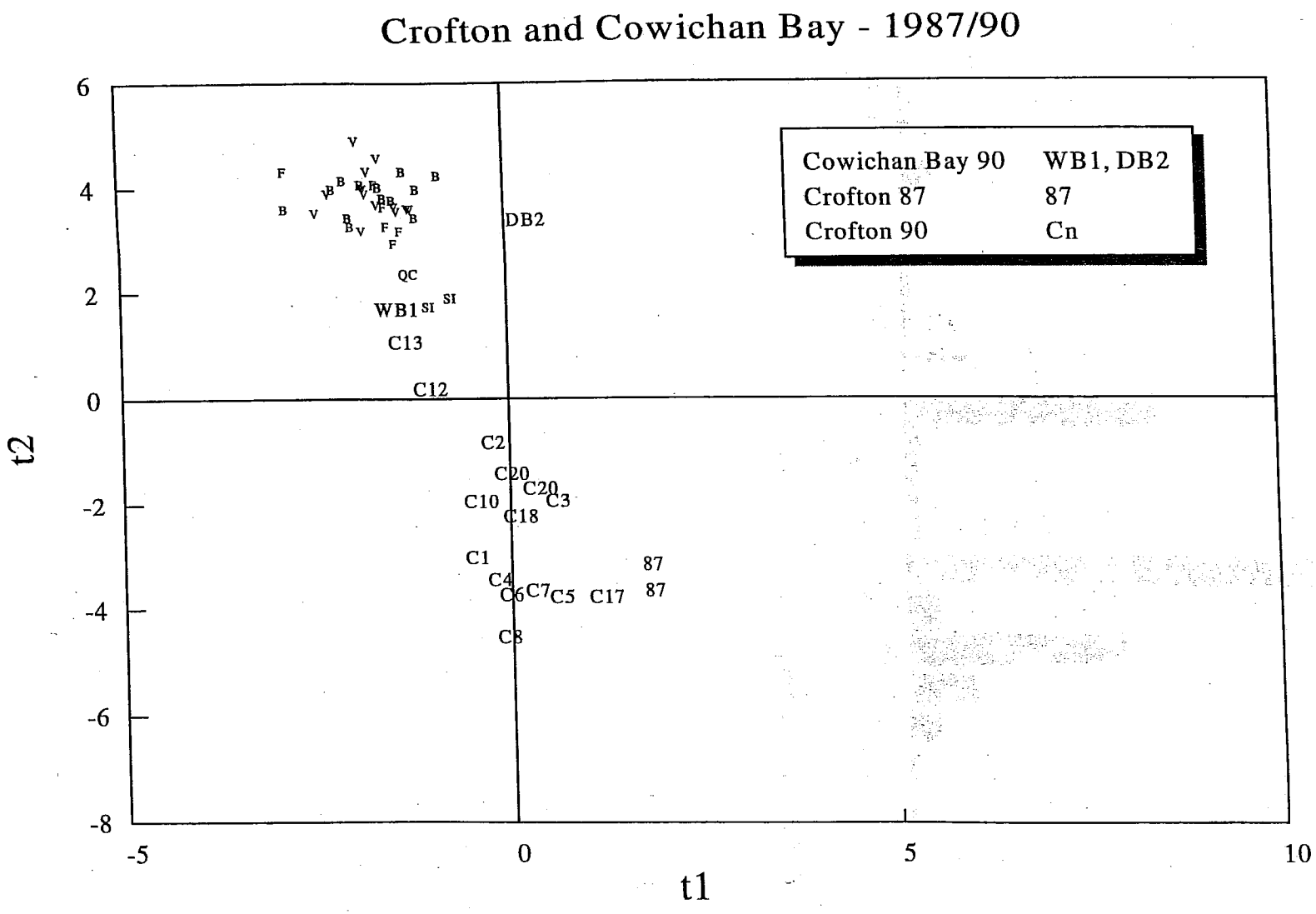


Figure 6-4. PCA projections for samples from Crofton in 1987 and 1990 and Cowichan Bay in 1990.

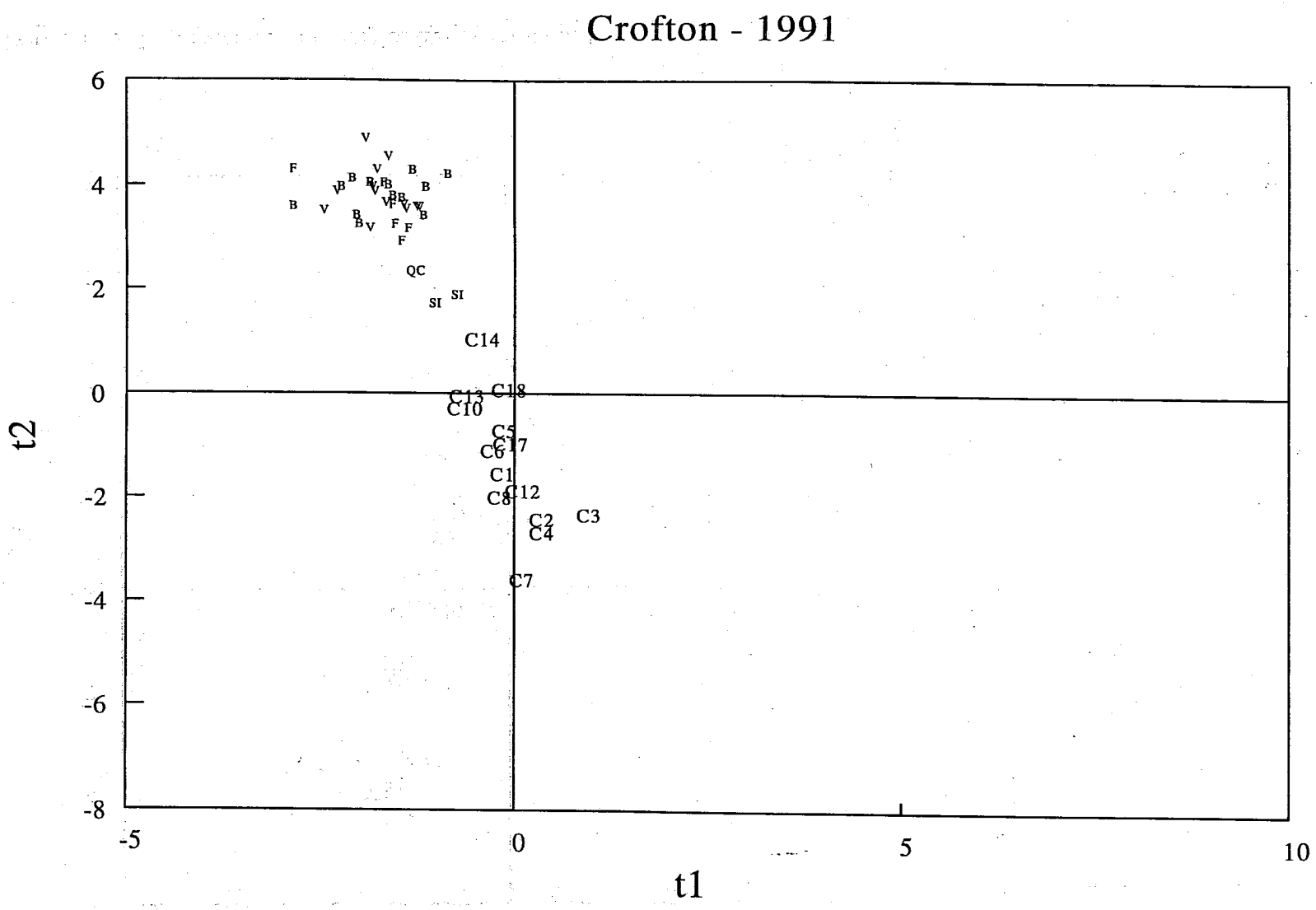


Figure 6-5. PCA projections for samples from Crofton in 1991.

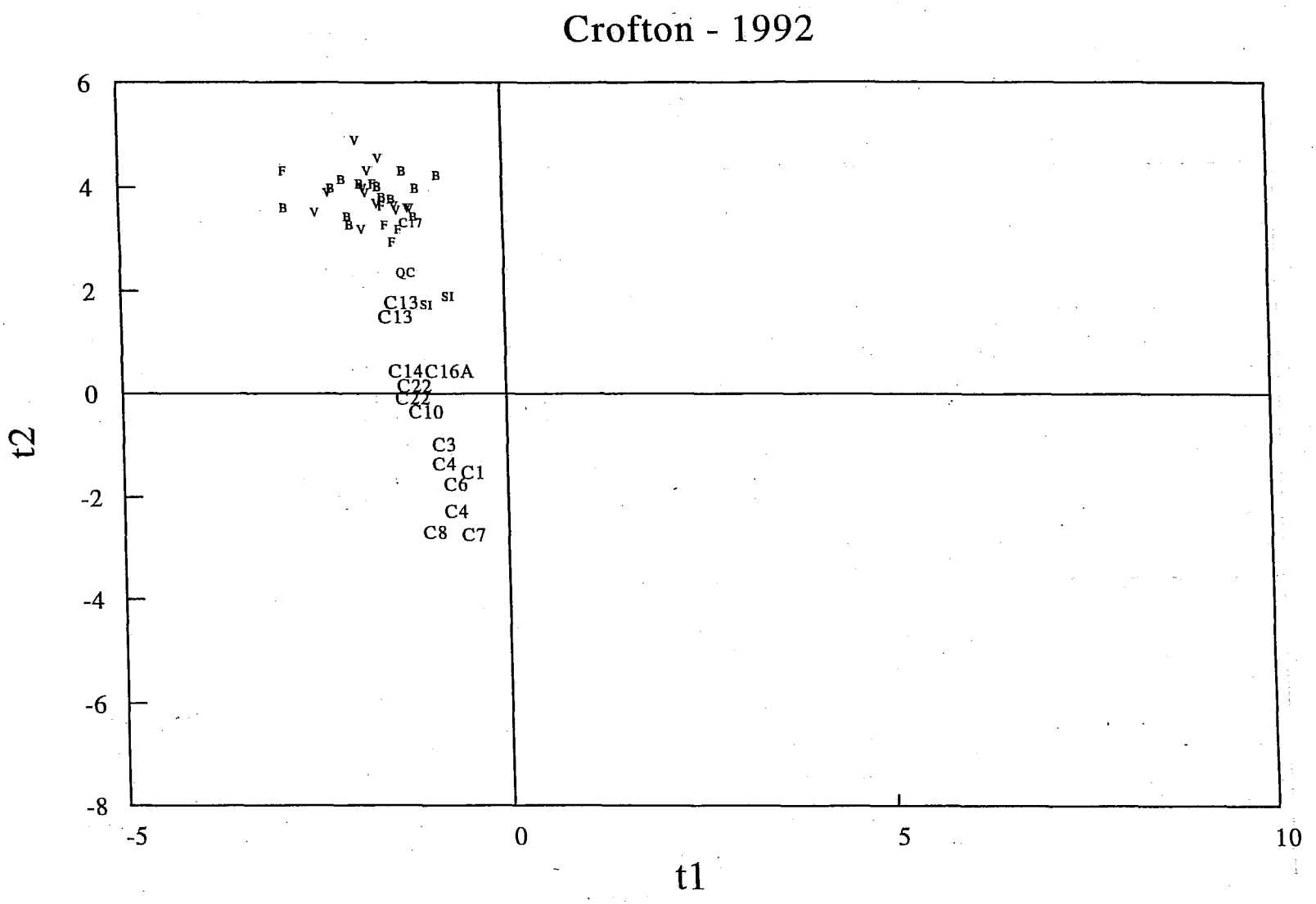


Figure 6-6. PCA projections for samples from Crofton in 1992.

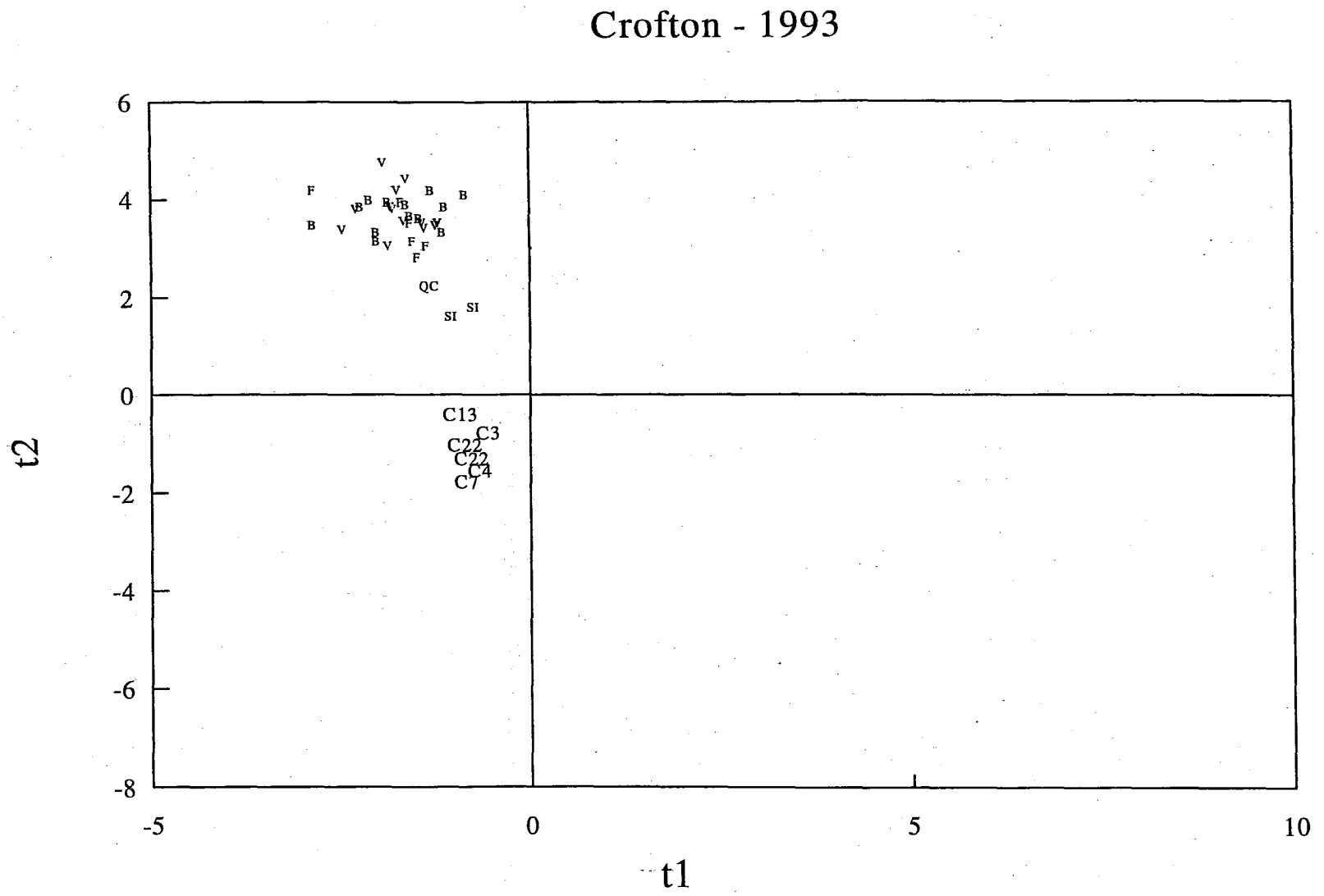


Figure 6-7. PCA projections for samples from Crofton in 1993.

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7. CAMPBELL RIVER

Two mills are located near to Campbell River: the Fletcher Challenge Canada Limited Elk Falls Pulp and Paper mill and the Campbell River Mills Limited Raven Lumber sawmill (Map 7-1). These mills will be referred to as the Elk Falls and Raven Lumber mills, respectively.

Crab samples were collected in the Campbell River area by Hatfield Consultants Ltd. in 1990, with most samples collected in January and a few supplementary samples collected in August and September 1990. From 1991 to 1993 mill monitoring program samples were collected annually by Hatfield Consultants Ltd. in February or March (Map 7-1). Axys Analytical Ltd. analyzed all Campbell River samples from 1990 to 1993. Crab samples from the Campbell River area had the highest proportion of non-Dungeness species of any area. In 1990 Red Rock crabs were obtained at Stations C2, C10 and C11. In 1991 and 1992 all samples were Dungeness crab except for Box crab from Station C9 in 1992. In 1993 Box crabs were obtained again at Station C9, as well as Stations C15 and C17A, while Kelp crab was sampled at Station C17B.

The Campbell River area is also notable for the number of samples with distinct congener profiles and PCA projections. Crab sample C7 from 1990 has a composition very different from all other crab samples from the Campbell River area. The sample contains high proportions of TCDFs; its congener profile and PCA projection (Figure 7-1 and Figure 7-4) is more reminiscent of the Gold River samples (Section 8) than of the Campbell River samples. By including this sample in the 1990 congener total, TEQ average and congener profile (Table 7-1; Figure 7-1 and Figure 7-4), the contribution of the TCDFs and the magnitude of the TEQ may be overestimated. However, because this sample may be indicative of pre-1990 discharges from the Elk Falls mill, it has been retained in the 1990 averages.

The Raven Lumber crab sample CR-C1 from 1990 and the Elk Falls mill monitoring program crab samples collected in 1990, 1991 and 1993 from Station C4 had PCA projections similar to that of the harbours (Section 4; Figure 7-4 to Figure 7-7). Because these samples were located close to the mills, they very likely had been influenced by pentachlorophenol wood preservative from contaminated wood chips. Samples C3 from 1990, C4 from 1992 and C7 from 1992 and 1993 had PCA projections slightly further removed from the harbour samples; these samples most likely had also been exposed to contaminated wood chips. Accordingly, these samples were not included in the congener total and TEQ averages for the Elk Falls mill.

If only Dungeness crab samples are considered, and if the unusual samples enumerated above are excluded, the PCA projections of the two Dungeness crab samples C1 and C5 may be considered to be representative of the pulp mill profile in 1990 (Figure 7-4). The 1990 congener total and calculated TEQ for just these two samples is only slightly different from the totals that include C7 (Table 7-1). Relative to a 1990 baseline, the Dungeness crab samples collected between Quadra Island and Vancouver Island in general exhibit only small changes in PCA composition and TEQ between 1991 and 1993 (Figure 7-5 to Figure 7-7; Table 7-1). A good example is

Station C2, located on Quadra Island directly across from the Elk Falls mill, which exhibits almost no change in PCA projection from 1991 to 1993.

From the PCA results it is evident that PCDD/Fs with a pulp mill composition were present in crab samples from Kanish Bay (C13) in 1992 and 1993 and from the north end of Quadra Island (C19, C20) in 1993 (Figure 7-6 and Figure 7-7). These samples exhibit a composition pattern that is indistinguishable from the pattern of Stations C1, C2, C5, C10 and C12. Samples from Station C9 on the southeast corner of Quadra Island (Francisco Point) are more difficult to categorize. The change in PCA projection between 1991 and 1992 at Station C9 could be due to the species change from Dungeness crab to Box crab, but sample C7 consisted of Dungeness crab in both years and went through a similar shift (Figure 7-5 and Figure 7-6). There was no further change in the composition of the two Box crab samples collected at Station C9 in 1992 and 1993. When considered in the context of a relatively constant mill-related PCA composition, it is likely that the Box crab samples from Station C9 and Mitlenatch Island (C17A) in 1993 reflect PCDD/Fs with a pulp mill origin.

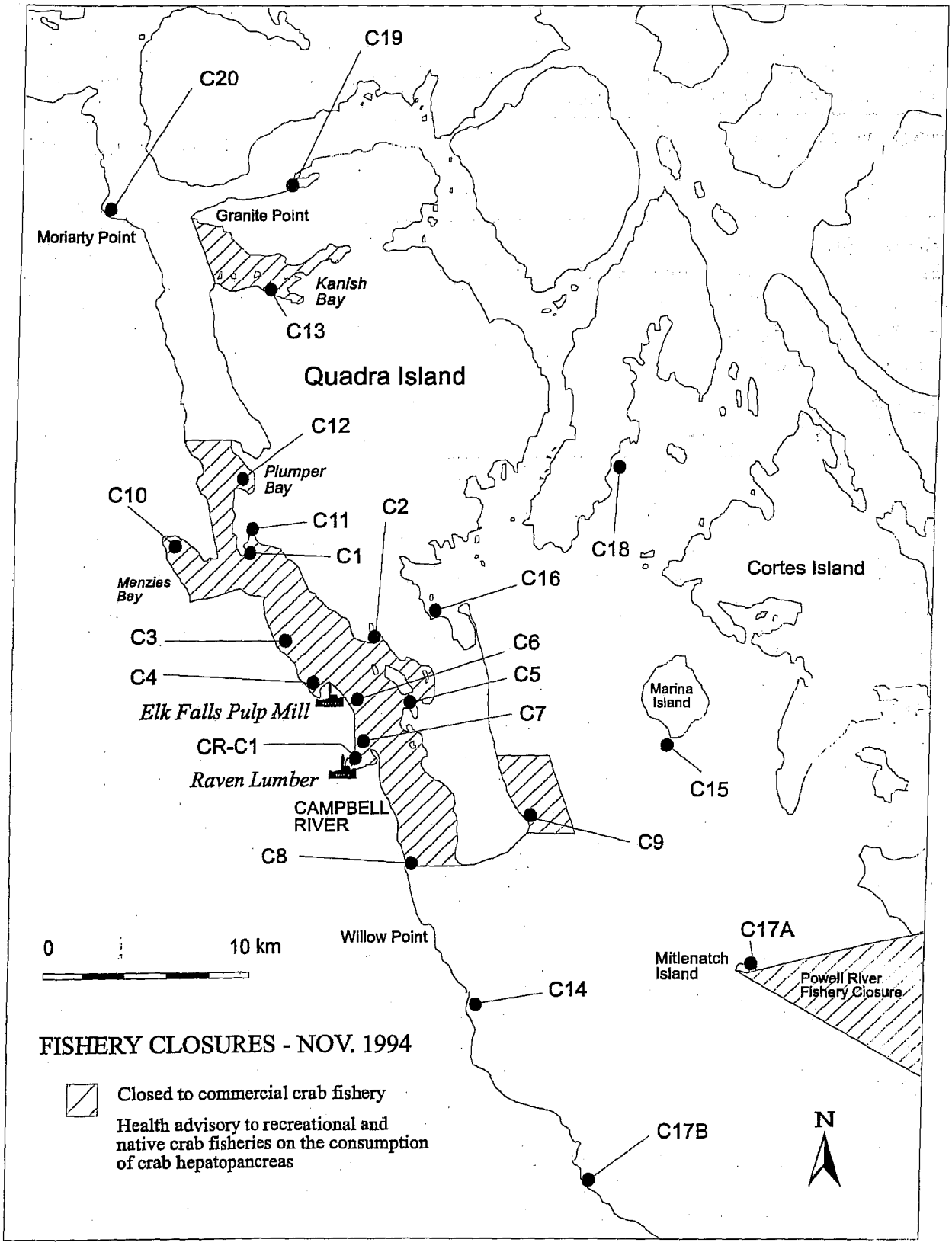
Sample C15 from Marina Island, on the east side of Quadra Island, had a composition identical to the pulp mill samples in 1992, but had a composition intermediate between the mill samples and harbour samples in 1993 (Figure 7-6 and Figure 7-7). The composition change between years may be due to a species change from Dungeness crab to Box crab. The 1992 Dungeness crab sample from Station C16 in Heriot Bay also projected between the mill samples and harbour samples and has apparently received PCDD/Fs from non-mill sources.

Sample C17B consisted of Kelp crabs from Oyster River, which is located south of Quadra Island along the coast of Vancouver Island. The TEQ for this sample (0.3 pg/g) was much lower than any of the TEQ values for the samples of Dungeness crab or Box crab. Because this is the only sample of kelp crab in the data set, no conclusions can be reached about the PCA projection of this sample.

Since a comparison of trends in TEQ data is questionable when different species are involved, the collection and analysis of crab species other than Dungeness crab is of dubious value in the current mill monitoring program. It is recommended that an effort be made to relocate Elk Falls sampling stations that are located to the east and south of Quadra Island (C9, C15 and C17) to nearby areas where the habitat is suitable for the collection of Dungeness crabs. Because Heriot Bay (C16) may have received PCDD/Fs from non-mill sources it is unsuitable as a substitute for C9.

Table 7-1. Dungeness crab hepatopancreas congener total and TEQ averages for samples with similar PCA projections from Campbell River.

Samples Averaged	**Congener Total**	**Calculated TEQ**
	pg/g wet weight	
Earliest Samples		
Elk Falls C1, C5 and C7, 1990 (n=3)	1240	100
Elk Falls C1 and C5, 1990 (n=2)	1200	90
Elk Falls C7, 1990 (n=1)	1300	120
Raven Lumber CR-C1, 1990 (n=1)	3900	130
Most Recent Samples		
Elk Falls C2 and C12, 1993 (n=3)	800	53



Map 7-1. Crab sampling locations near Campbell River.

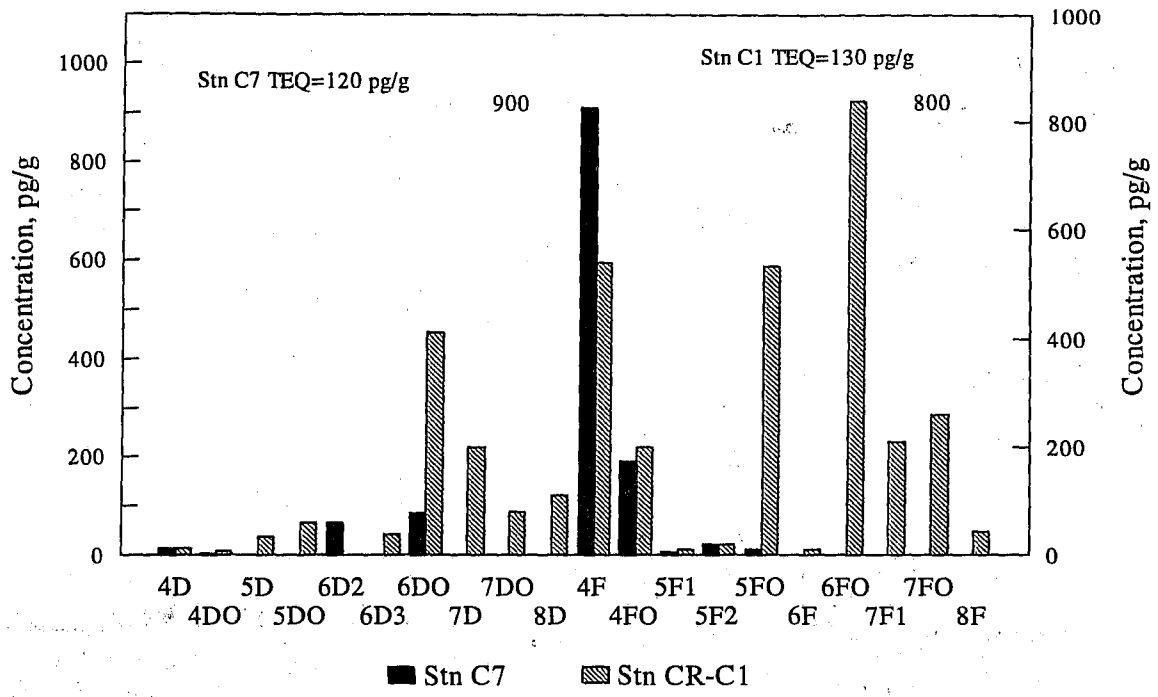


Figure 7-1. Congener profiles for stations in the Campbell River estuary in 1990 (n=1 for both).

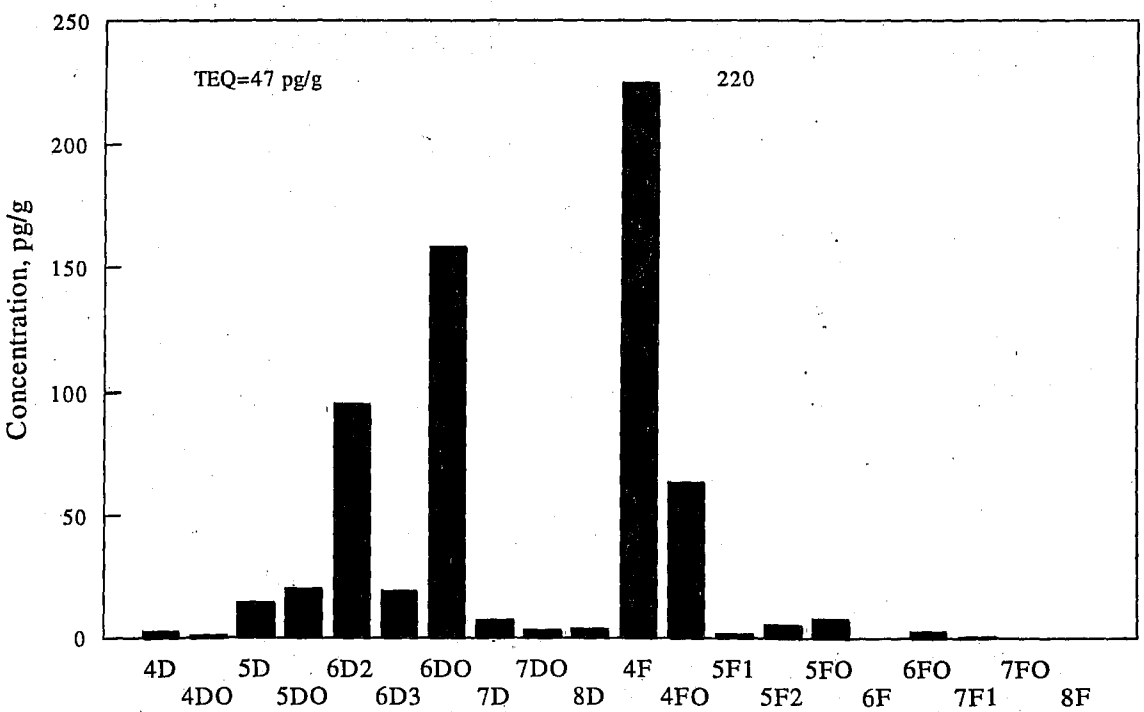


Figure 7-2. Campbell River congener profiles for 1990 (n=3).

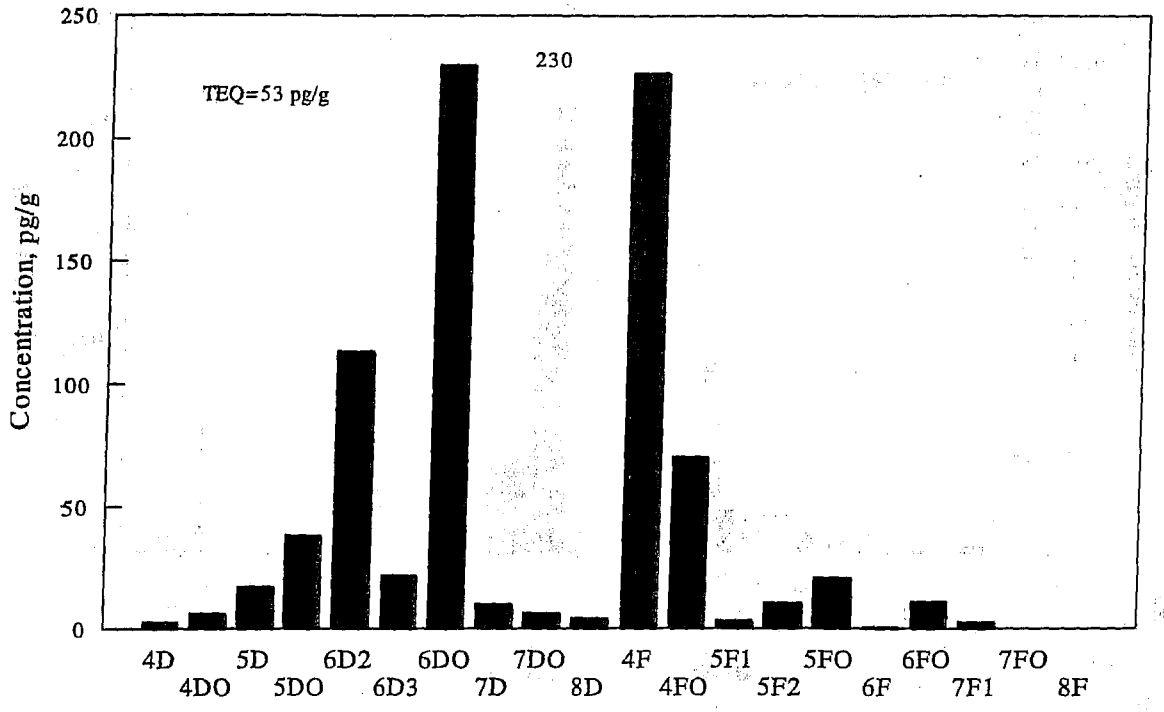


Figure 7-3. Campbell River congener profiles for 1993 (n=3).

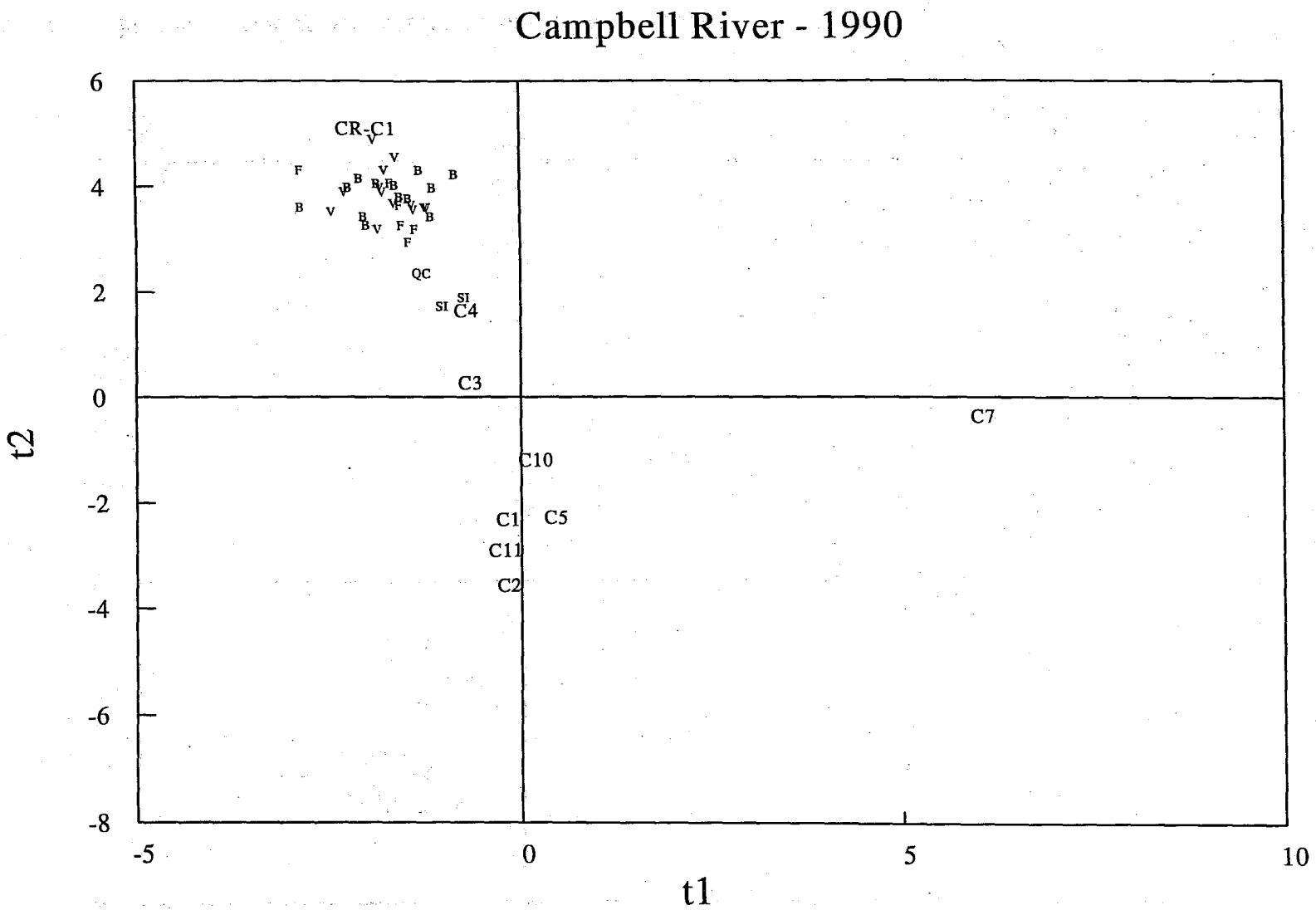


Figure 7-4. PCA projections for samples from Campbell River in 1990.

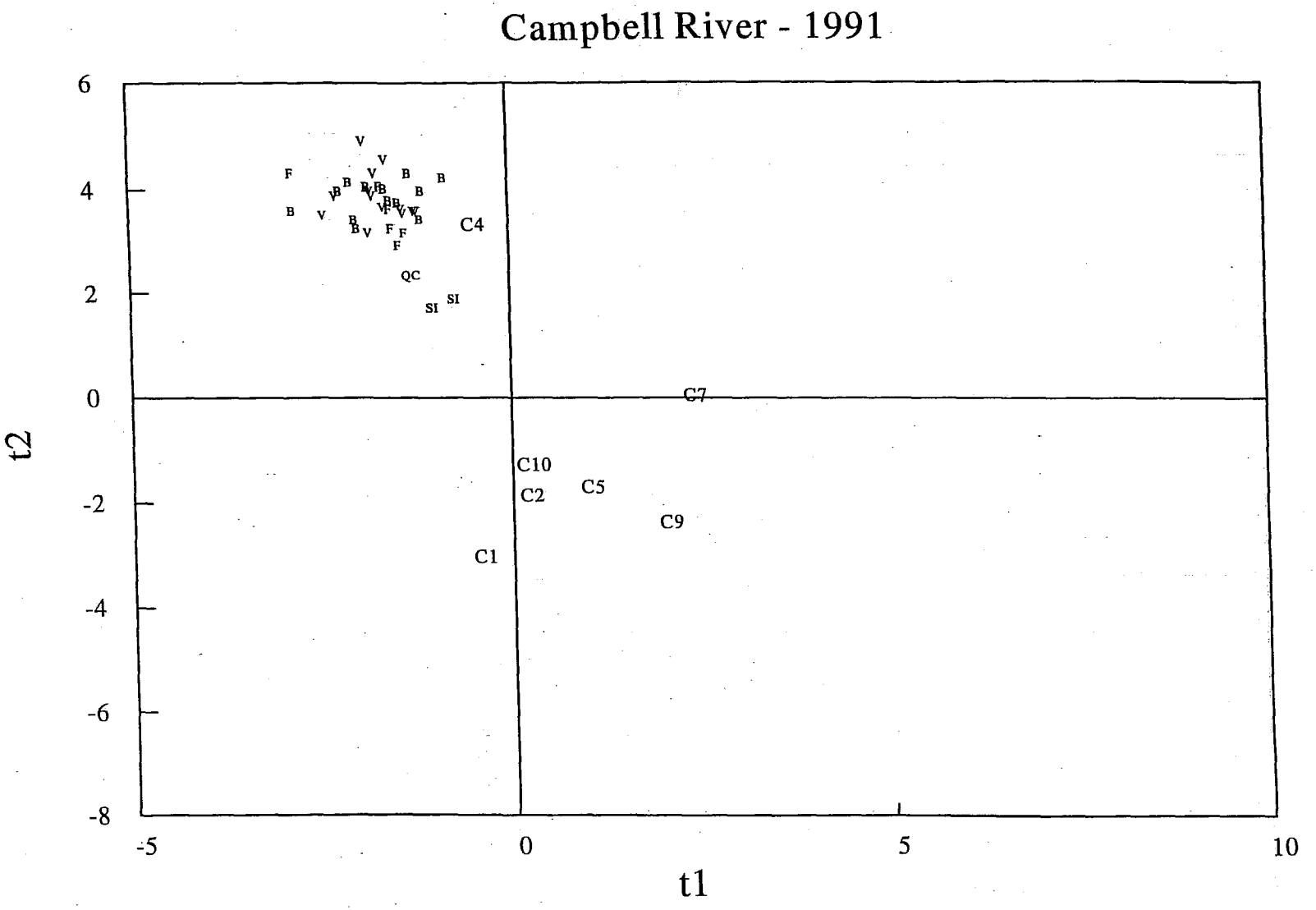


Figure 7-5. PCA projections for samples from Campbell River in 1991.

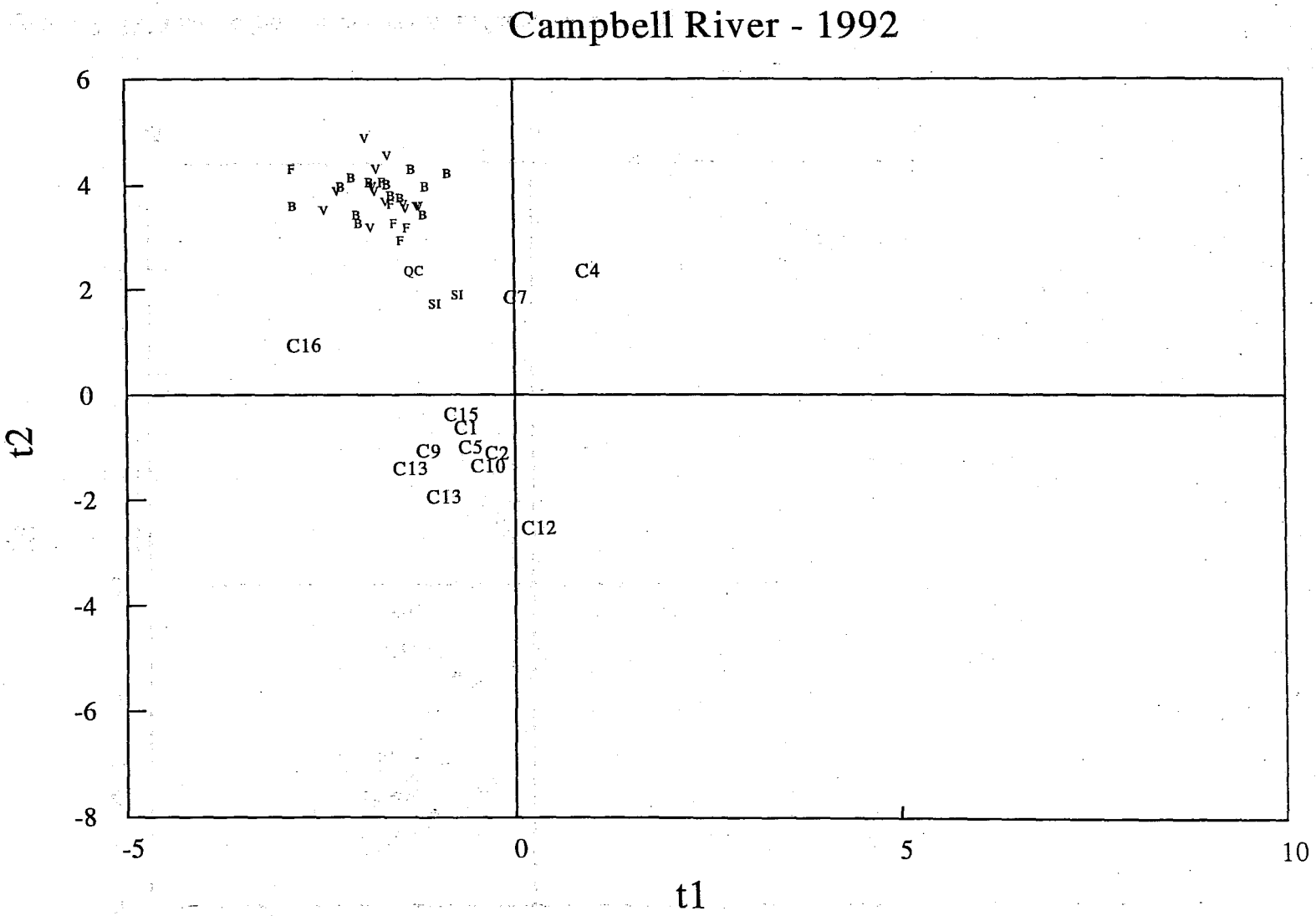


Figure 7-6. PCA projections for samples from Campbell River in 1992.

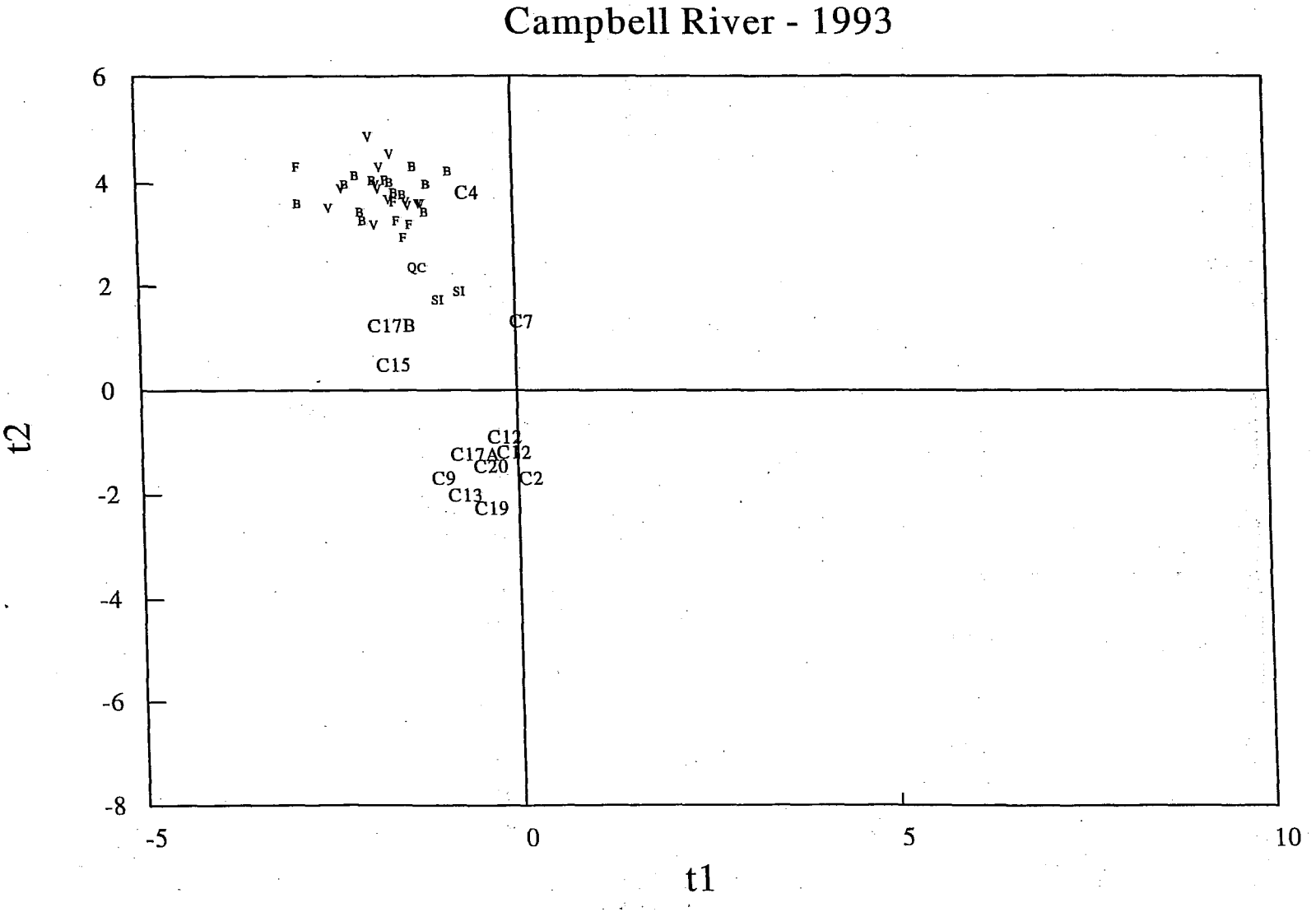


Figure 7-7. PCA projections for samples from Campbell River in 1993.

8. GOLD RIVER

Crab samples were collected in the vicinity of the Canadian Pacific Forest Products Limited (now Avenor Inc.) Gold River Pulp Mill by Beak Consultants Ltd. in 1990, with most samples collected in January and February and a few supplementary samples collected in September and October. Subsequent mill monitoring program samples were collected by Beak Consultants Ltd. in March 1991, February 1992 and April 1993 (Map 8-1). Axys Analytical Ltd. analyzed all Gold River mill monitoring program samples from 1990 to 1992. Wellington Laboratories Ltd. of Guelph Ontario analyzed the 1993 samples. In May 1993 an additional commercial fishery crab sample was collected in the Nootka Sound/Nutchaliz Inlet area by the Department of Fisheries and Oceans and analyzed by the Institute of Ocean Sciences. All samples from the Gold River area were Dungeness crab.

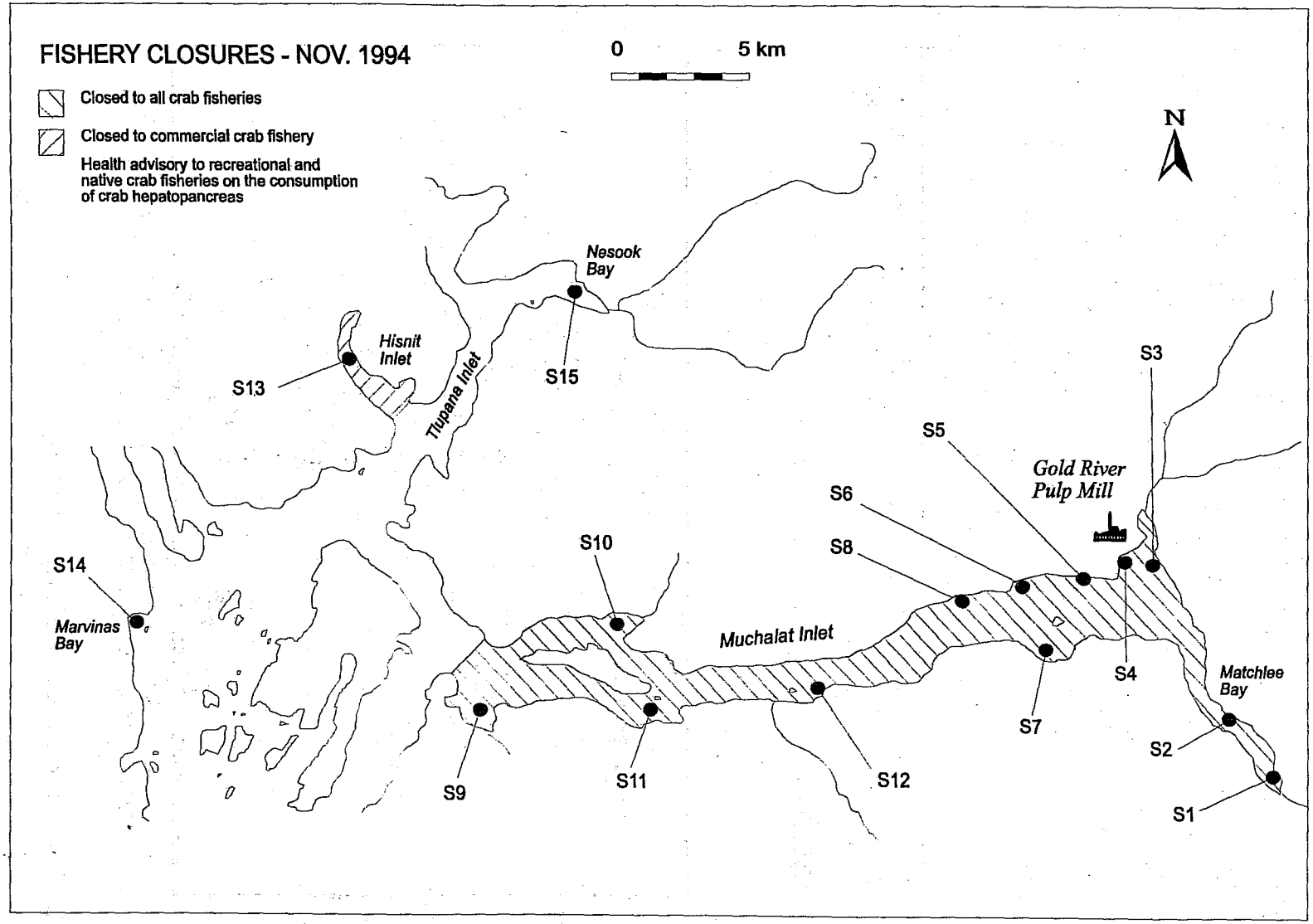
PCA projections and PCDD/F congener profiles for crab hepatopancras samples collected near to the Gold River mill in 1990 are unusual because the samples are made up almost entirely of TCDFs (Figure 3-3, Figure 8-1 and Figure 8-3). This is the composition that would be expected if very few pentachlorophenol contaminated chips had been used in the bleach plant at the mill (see Section 3).

PCA results indicate that PCDD/Fs with a pulp mill origin were present throughout Muchalat Inlet in 1990 (Figure 8-3 to Figure 8-6; Map 8-1). Samples collected outside the inlet through 1993 indicate that mill PCDD/Fs are present as far north as Netsook Bay and as far west as Marvinas Bay. The commercial fishery sample from outside the mill monitoring area whose location was recorded as Nootka Sound/Nutchaliz Inlet (Section 4) also has a congener profile and PCA projection that may indicate influence by discharges from the Gold River mill (Figure 4-8 and Figure 8-6).

Between 1990 and 1993 the calculated TEQ decreased by more than an order of magnitude for stations closest to the mill (Table 8-1). Congener profiles and PCA projections both indicated a decrease in the proportion of toxic congeners over this time (Figure 8-1, Figure 8-2, Figure 8-3 and Figure 8-6). The change in composition between years principally has been reflected as an increase in the HxCDDs. PCA projections remained relatively consistent within each year, which indicates that the change in congener composition has proceeded uniformly over time.

Table 8-1. Dungeness crab hepatopancreas congener total and TEQ averages for samples with similar PCA projections from Gold River.

Samples Averaged	**Congener Total**	**Calculated TEQ**
	pg/g wet weight	
Earliest Samples		
Gold River S3, S4, S5, S6 and S8, 1990 (n=5)	13,000	1100
Most Recent Samples		
Gold River S3, S5 and S6, 1993 (n=4)	1200	99
Nootka Sound, 1993 (n=1)	82	5.4



Map 8-1. Crab sampling locations near Gold River.

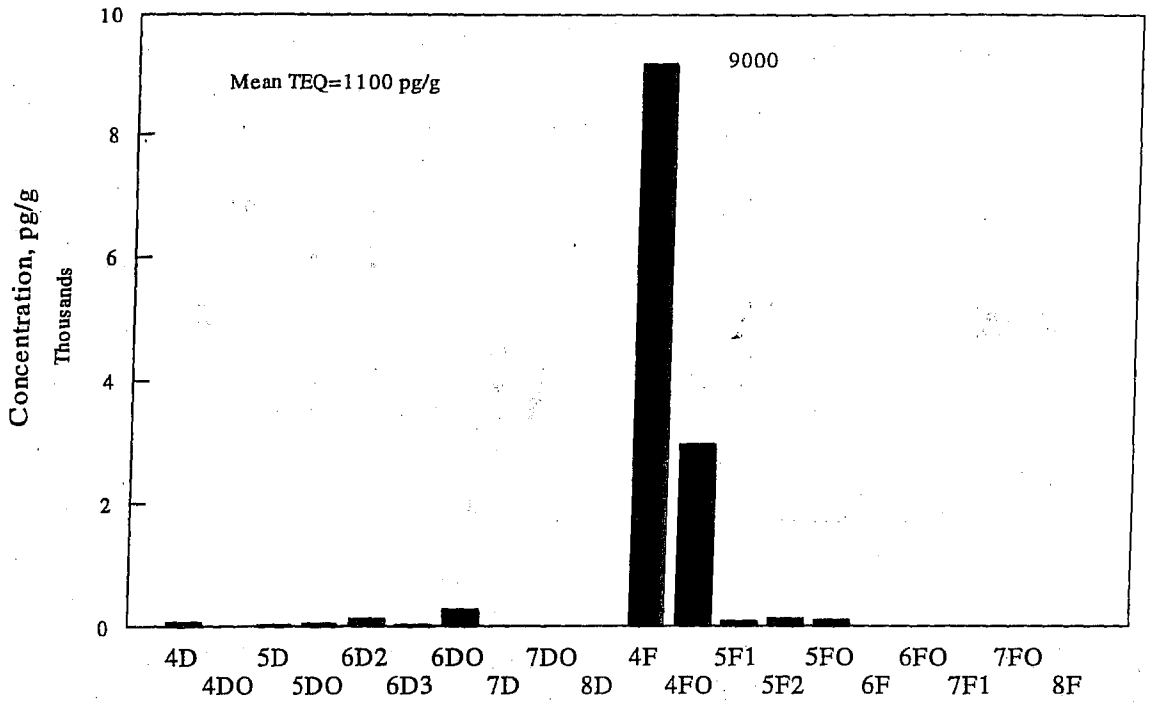


Figure 8-1. Gold River congener profiles for 1990 (n=5).

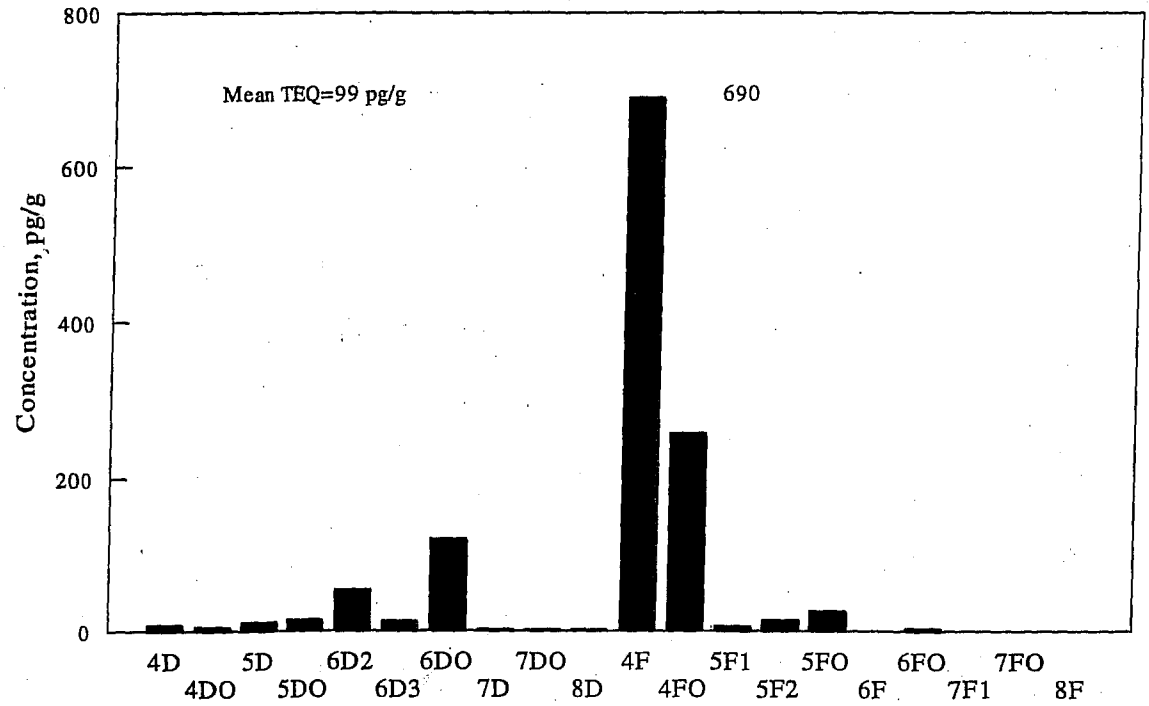


Figure 8-2. Gold River congener profiles for 1993 (n=4).

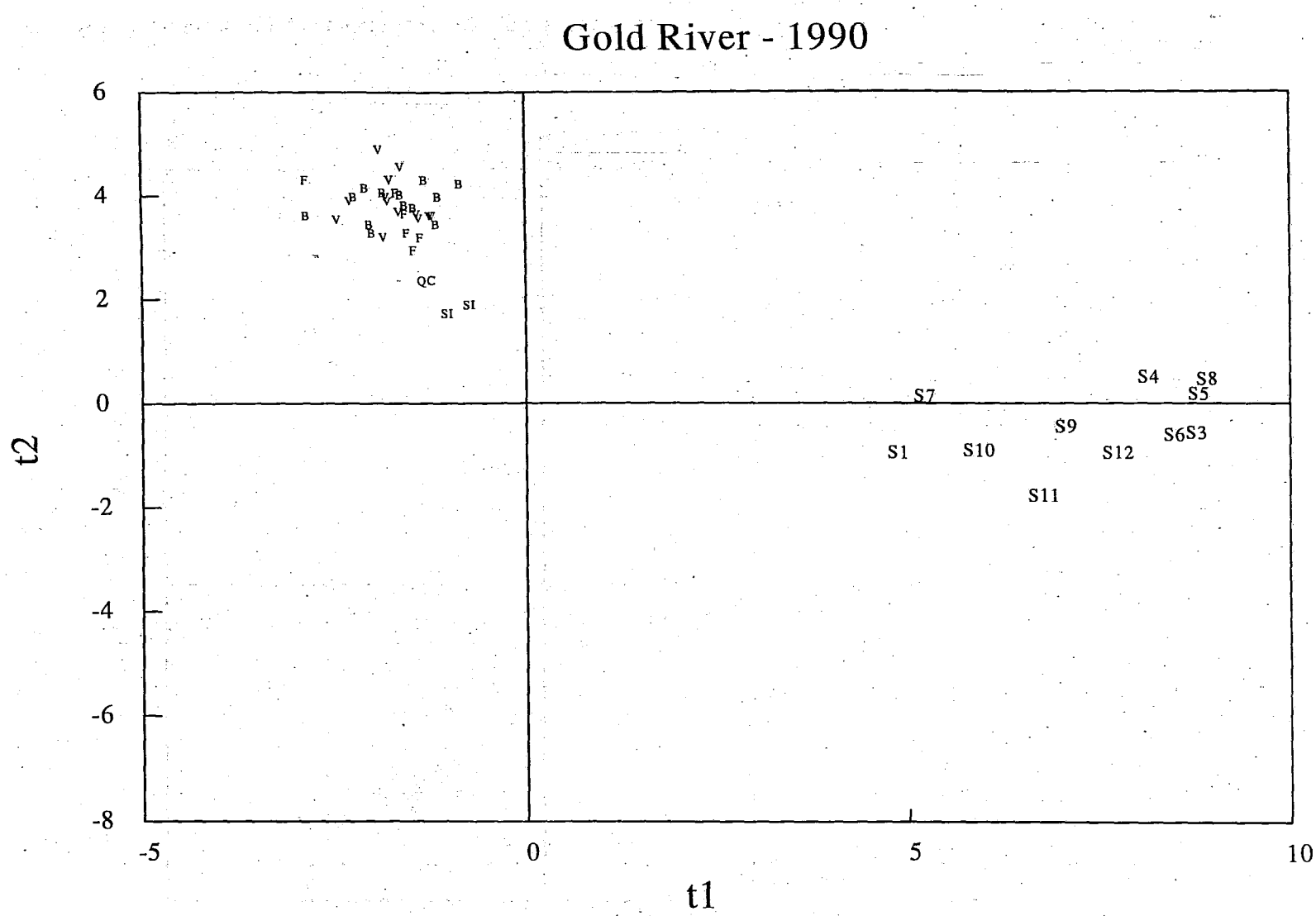


Figure 8-3. PCA projections for samples from Gold River in 1990.

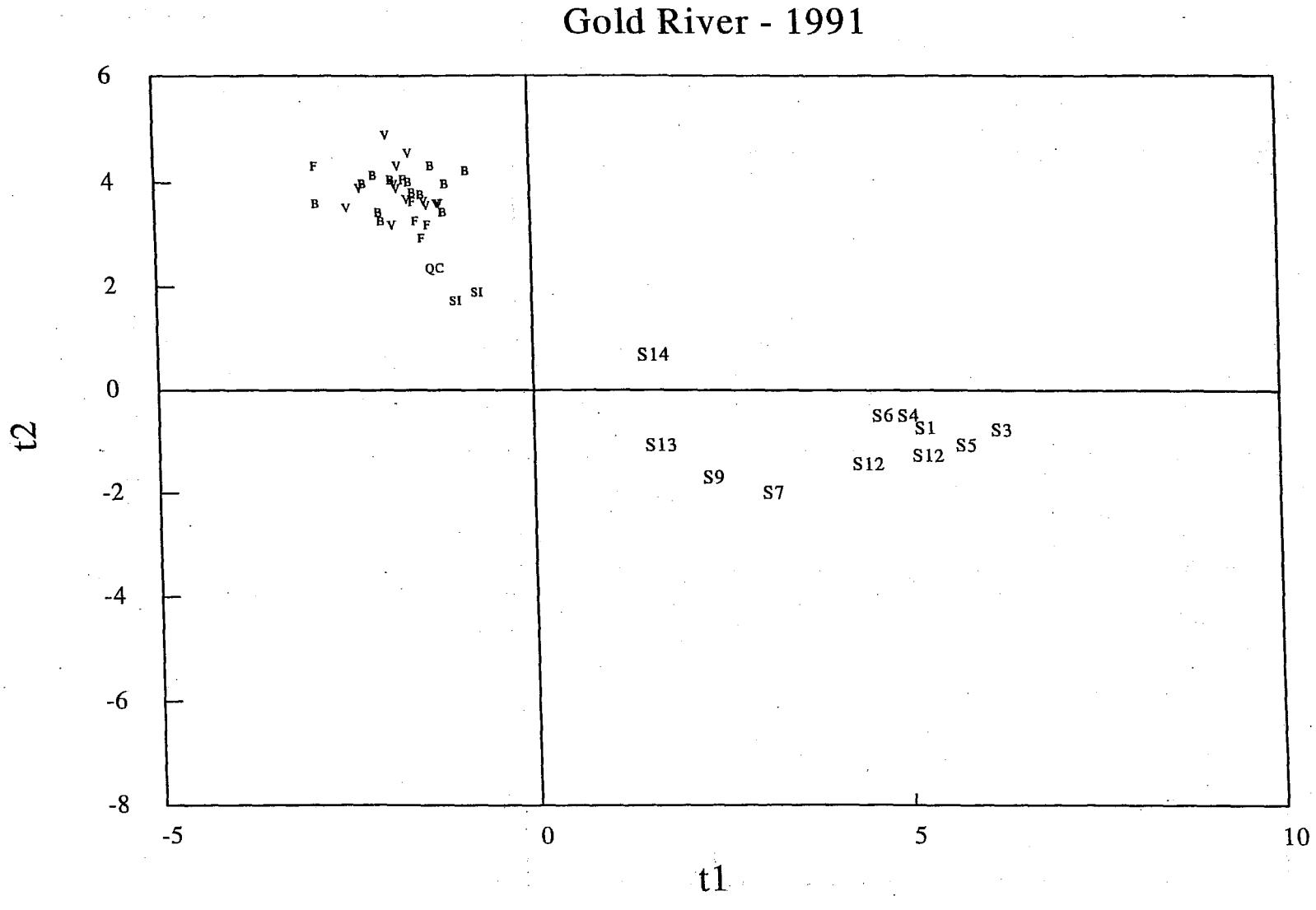


Figure 8-4. PCA projections for samples from Gold River in 1991.

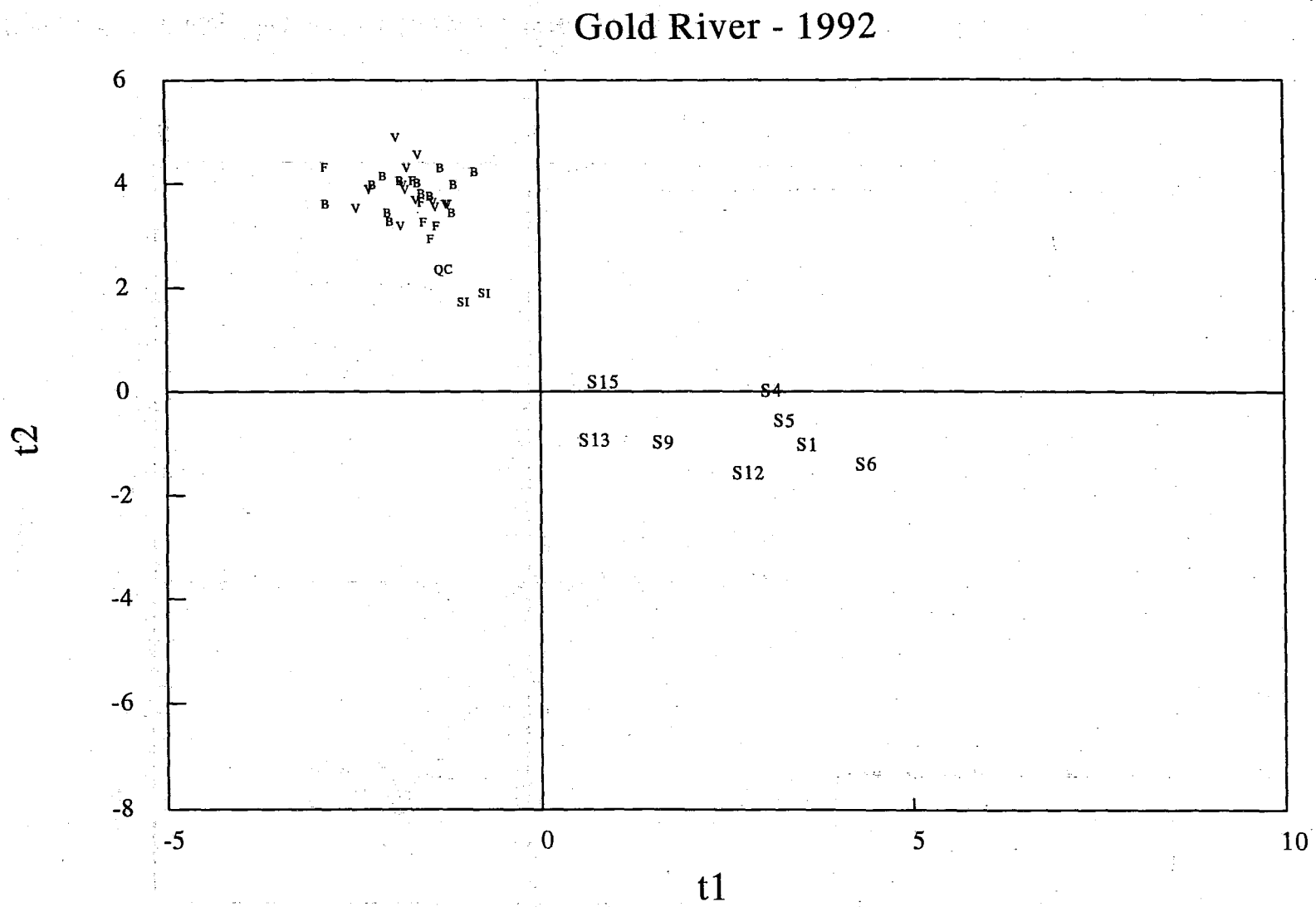


Figure 8-5. PCA projections for samples from Gold River in 1992.

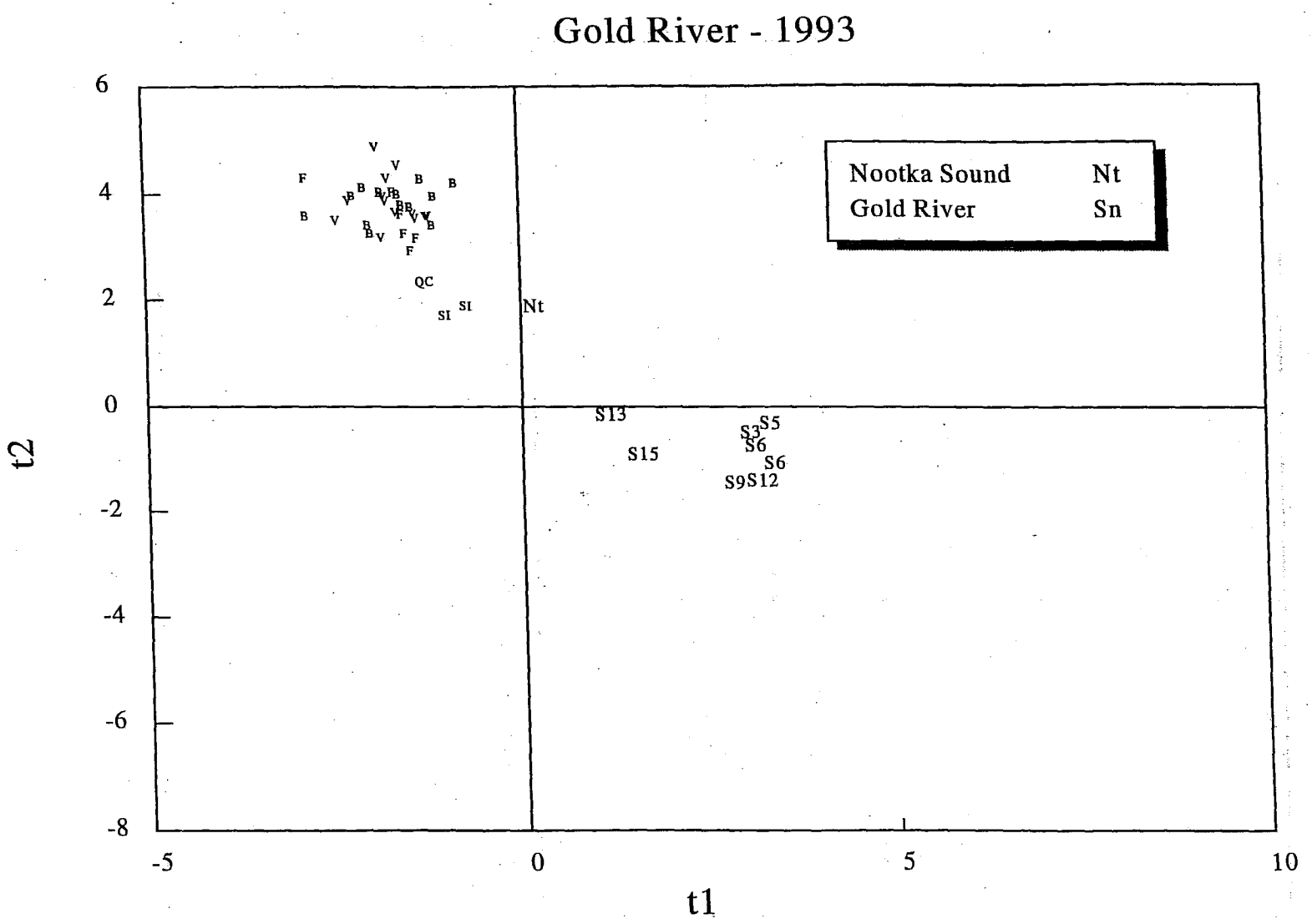


Figure 8-6. PCA projections for samples from Gold River in 1993.

9. NANAIMO

Crab samples were collected in Nanaimo Harbour and near the Harmac Pacific Inc. Pulp Mill by the Environmental Protection Service in July 1987 and analyzed by the Canadian Wildlife Service (Norstrom *et al.* 1988). Subsequent samples were collected in Nanaimo Harbour and near the Harmac mill by Hatfield Consultants Ltd. in January 1990 (Map 9-1). The sampling range was extended further to the north and to the south in August 1990; one of these samples (C10) had too few congeners detectable to be used in the PCA model (Table 2-2). Nanaimo and Harmac mill monitoring samples were then collected by Hatfield Consultants Ltd. in March 1991 and February 1992 and by G3 Consulting Ltd. in March 1993. Axys Analytical Ltd. analyzed all mill monitoring program samples from 1990 to 1992. Envirotest Laboratories Ltd. of Edmonton Alberta analyzed the 1993 samples. Crab species other than Dungeness crab have been obtained at a few locations in the Nanaimo area; Red Rock crabs were obtained at Stations C4 and C7 in 1990, at Station C4 in 1991 and at Station C5 in 1993.

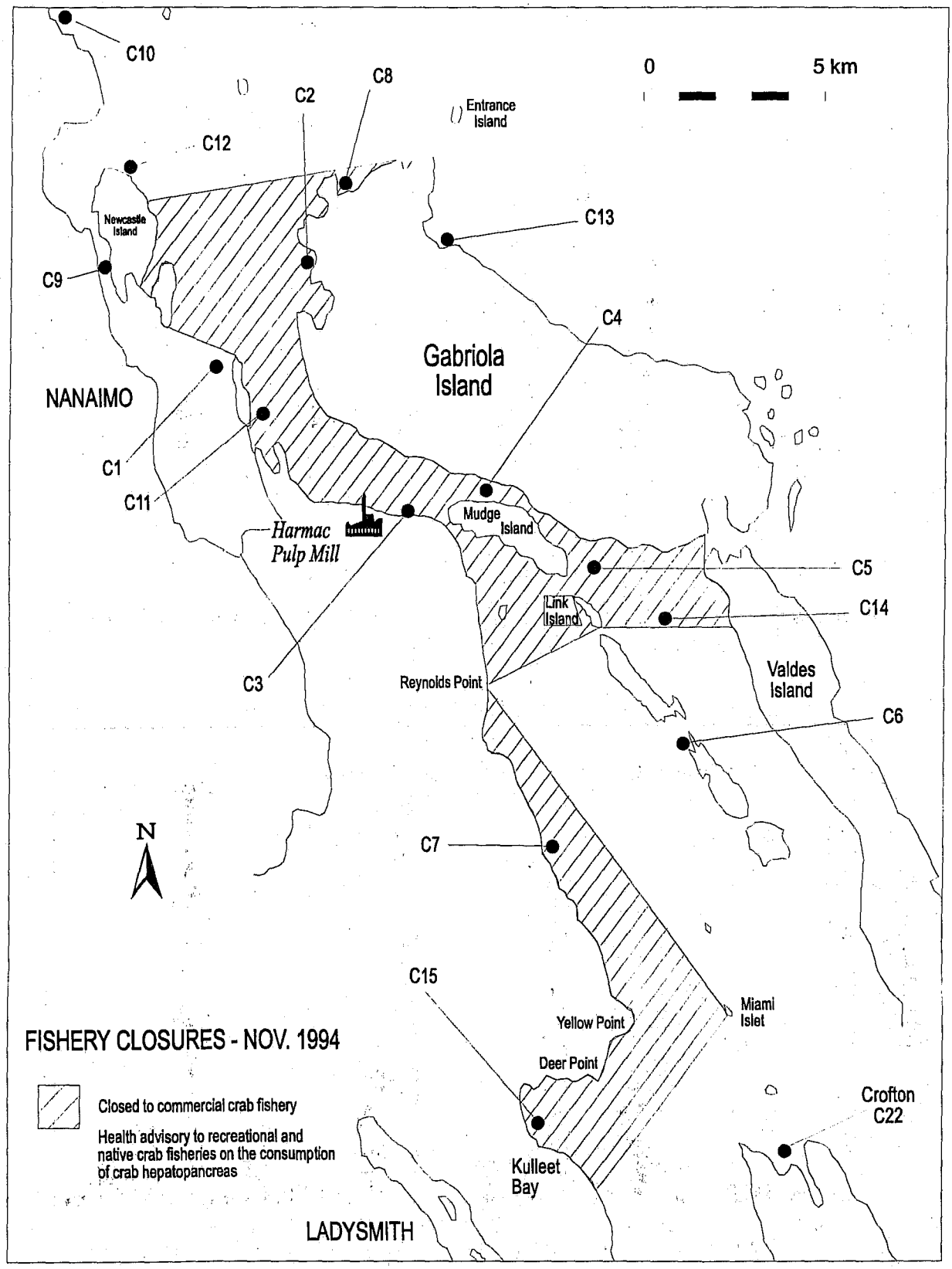
Interpretation of the PCA results for the Nanaimo area illustrates the different PCDD/F fingerprints of Nanaimo Harbour and of the Harmac mill and highlights the utility of PCA in ascribing a source to individual crab samples (Section 3; Figure 3-3 and Figure 3-4). Samples N87, C1 and C9 from Nanaimo Harbour project with the Victoria and Vancouver Harbour samples and change only slightly in composition profile between 1987 and 1992 (Figure 9-1, Figure 9-2; Figure 9-5 to Figure 9-7). Nanaimo Harbour samples have a slightly higher proportion of the TCDFs than the larger harbours; this suggests that chlorine bleaching effluents from the mill have also made a contribution. At a few locations from the boundary area just outside Nanaimo Harbour (C1 and C11; Map 9-1), crab samples had a harbour pattern in one year and a mill pattern in another year. Over the six years of sampling in the Nanaimo area, a smaller decrease in both concentration and TEQ has occurred in the harbour samples than in the mill samples (Table 9-1).

Crab samples from the vicinity of the Harmac mill project with mill samples from other areas and have a composition that has remained distinct from the harbours through to 1993 (Figure 9-3 and Figure 9-4; Figure 9-5 to Figure 9-8). Congener profiles and PCA projections for the earliest sampling times indicate that only a small change had occurred in congener composition between 1987 and January 1990. However, a progressive shift to the upper left in PCA projections, indicating a progressive increase in the HxCDD proportion, occurs for samples collected between 1990 and 1993. The change in composition has proceeded uniformly over time and congener compositions and PCA projections have remained relatively consistent in each year.

In 1993 crab samples ranging from Lock Bay (C13) in the north, on the outside of Gabriola Island, to the Harmac mill (C2), to Kulleet Bay (C15) in the south had the same congener composition (Figure 9-8; Map 9-1). Hence PCDD/Fs from the Harmac mill have dispersed and persisted throughout the region. In the south, the 1992 and 1993 PCA projections for Nanaimo/Harmac Station C15 overlap with Crofton Station C22 (see Section 6) — projections are consistent with either mill as a source or with mixed inputs.

Table 9-1. Dungeness crab hepatopancreas congener total and TEQ averages for samples with similar PCA projections from Nanaimo Harbour and the Harmac mill site.

Samples Averaged	**Congener Total**	**Calculated TEQ**
	pg/g wet weight	
Nanaimo Harbour — Earliest to Most Recent Samples		
Nanaimo Harbour, 1987 (n=1)	470	27
Nanaimo Harbour C1 and C9, 1990 (n=2)	300	15
Nanaimo Harbour, 1987 (n=1); C1 and C9, 1990 (n=2)	360	19
Nanaimo Harbour C1, C9 and C12, 1992 (n=4)	360	18
Harmac Pulp Mill — Earliest to Most Recent Samples		
Harmac Mill, 1987 (n=1)	1100	82
Harmac Mill C2, C3, C5, C6 and C8, 1990 (n=5)	810	65
Harmac Mill, 1987 (n=1); C2, C3, C5, C6 and C8, 1990 (n=5)	860	68
Harmac Mill C2 and C3, 1993 (n=3)	290	14



Map 9-1. Crab sample locations near Nanaimo.

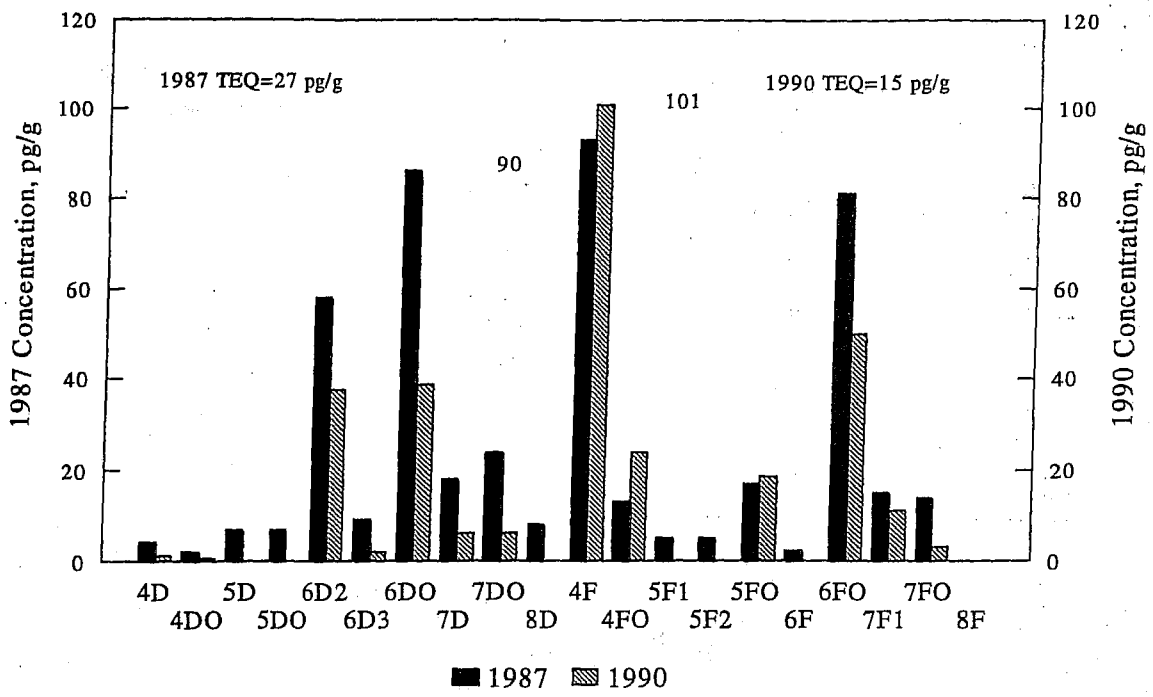


Figure 9-1. Nanaimo Harbour congener profiles for 1987 (n=1) and 1990 (n=2).

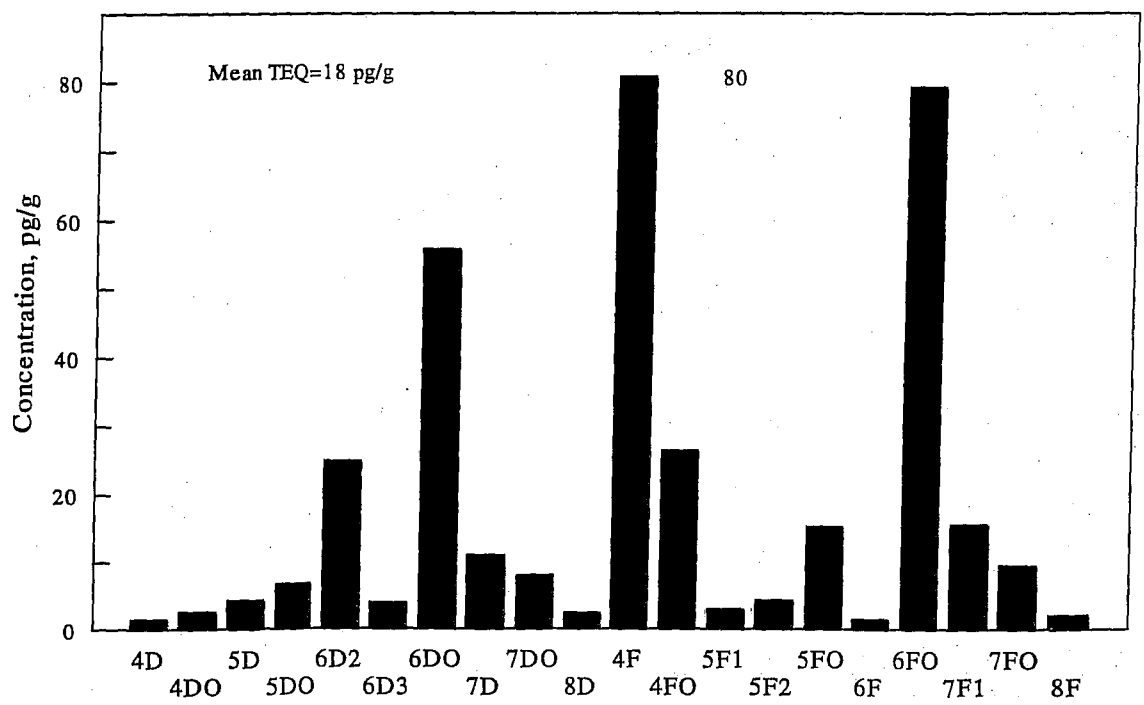


Figure 9-2 Nanaimo Harbour congener profiles for 1992 (n=4).

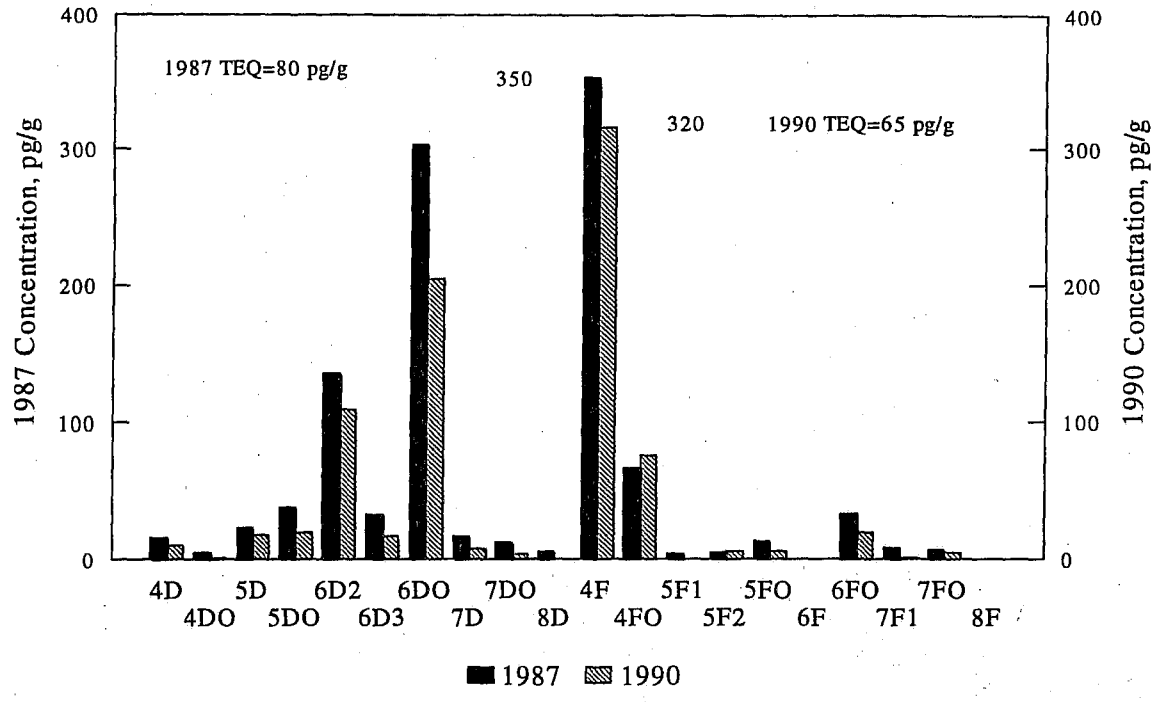


Figure 9-3. Nanaimo/Harmac mill congener profiles for 1987 (n=1) and 1990 (n=5).

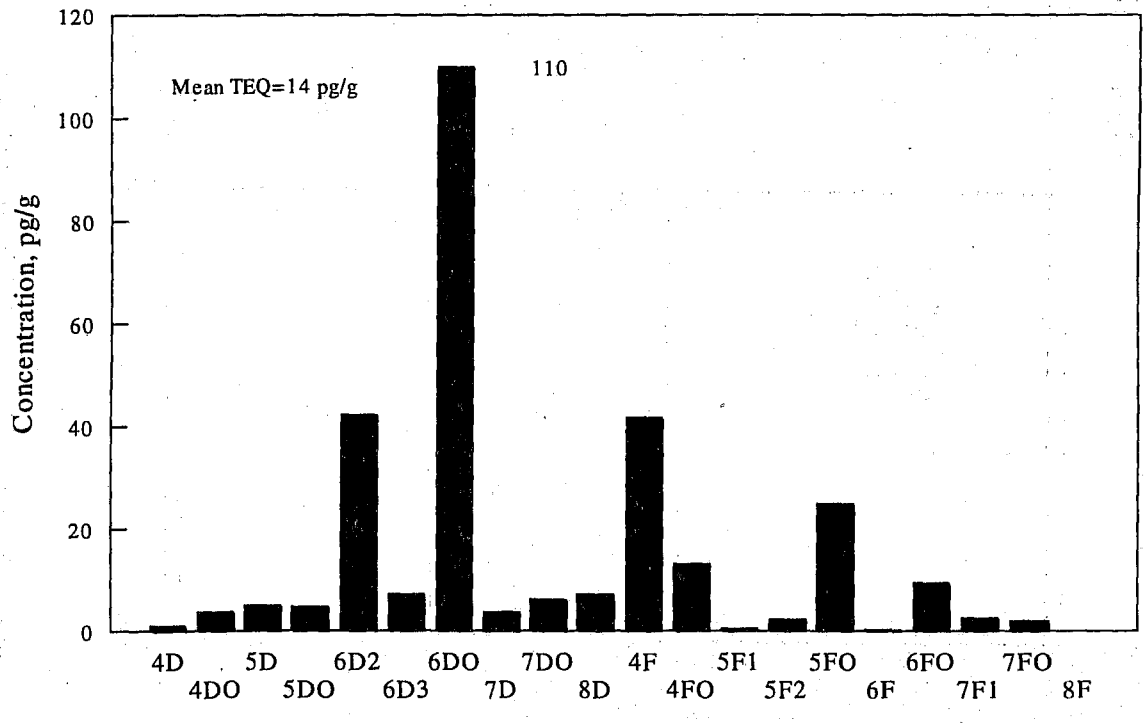


Figure 9-4. Nanaimo/Harmac mill congener profiles for 1993 (n=3).

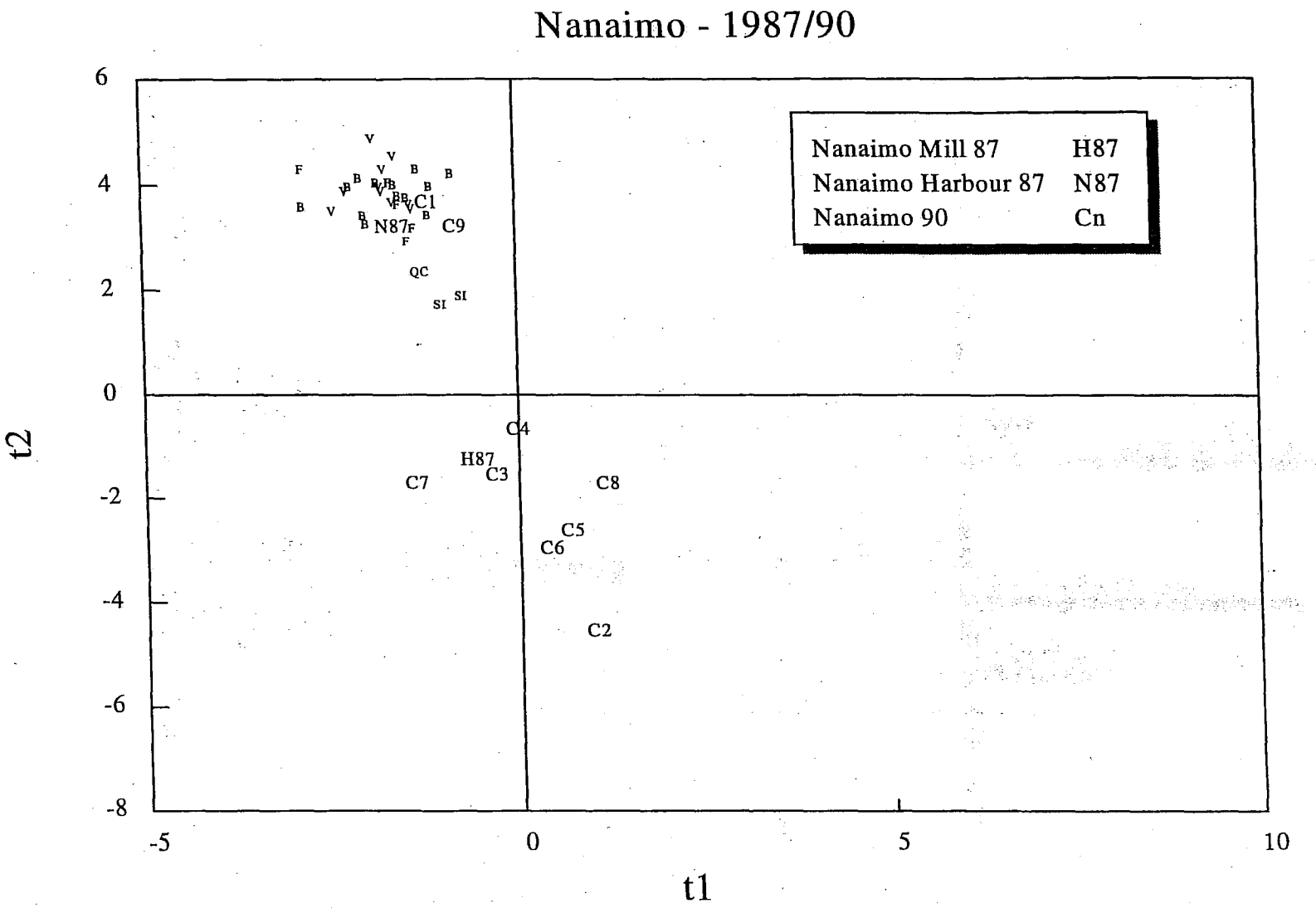


Figure 9-5. PCA projections for samples from Nanaimo in 1990.

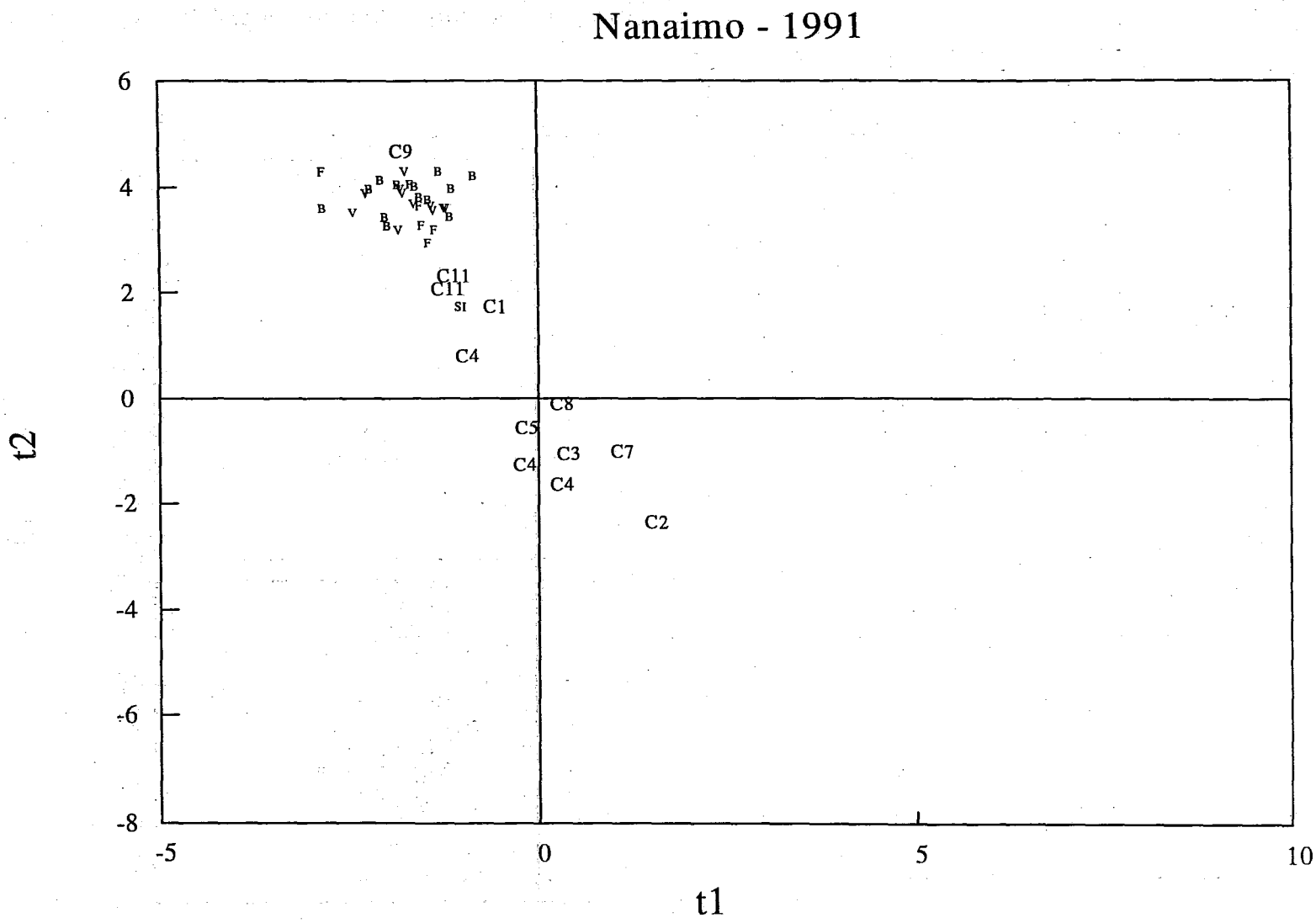


Figure 9-6. PCA projections for samples from Nanaimo in 1991.

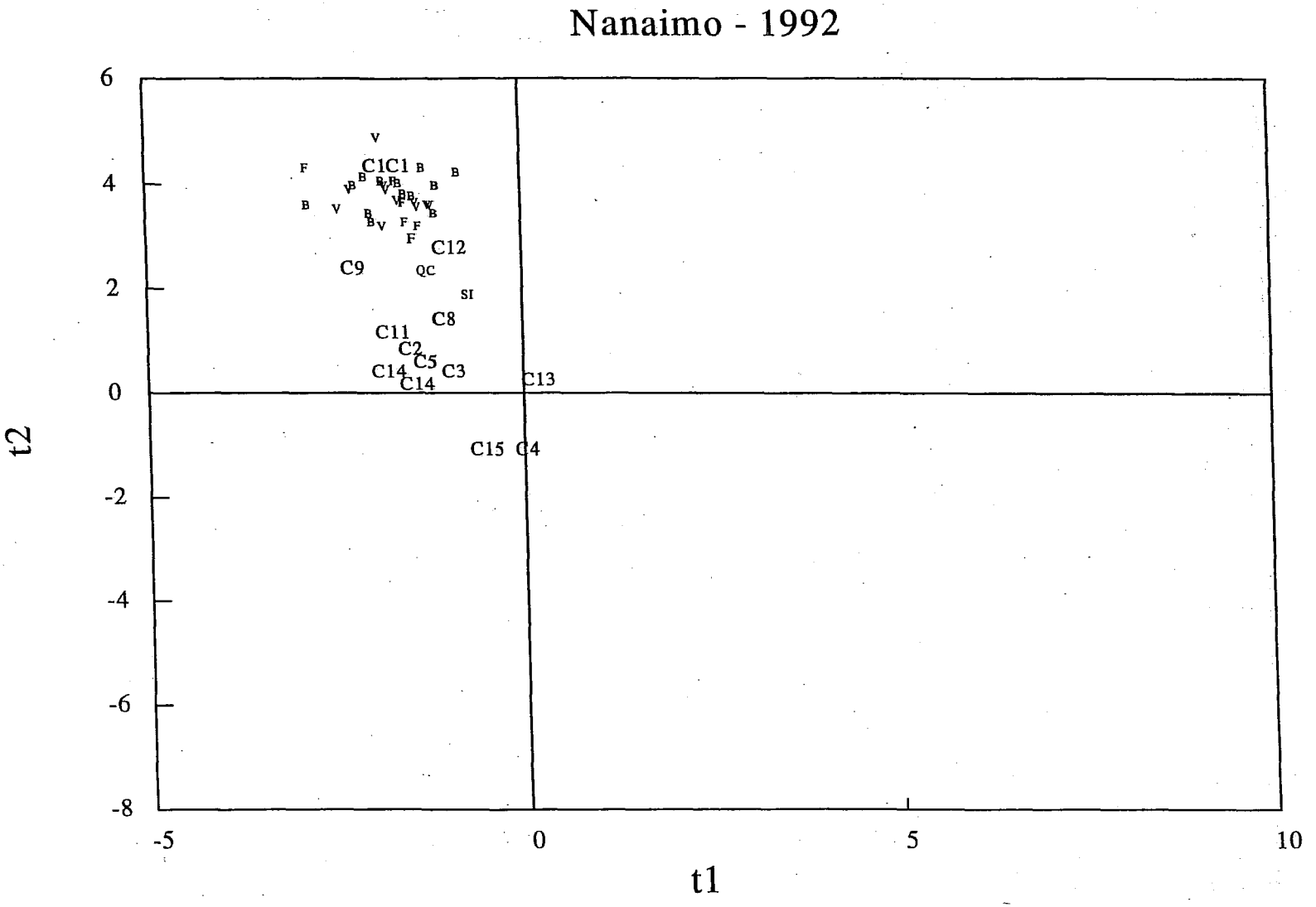


Figure 9-7. PCA projections for samples from Nanaimo in 1992.

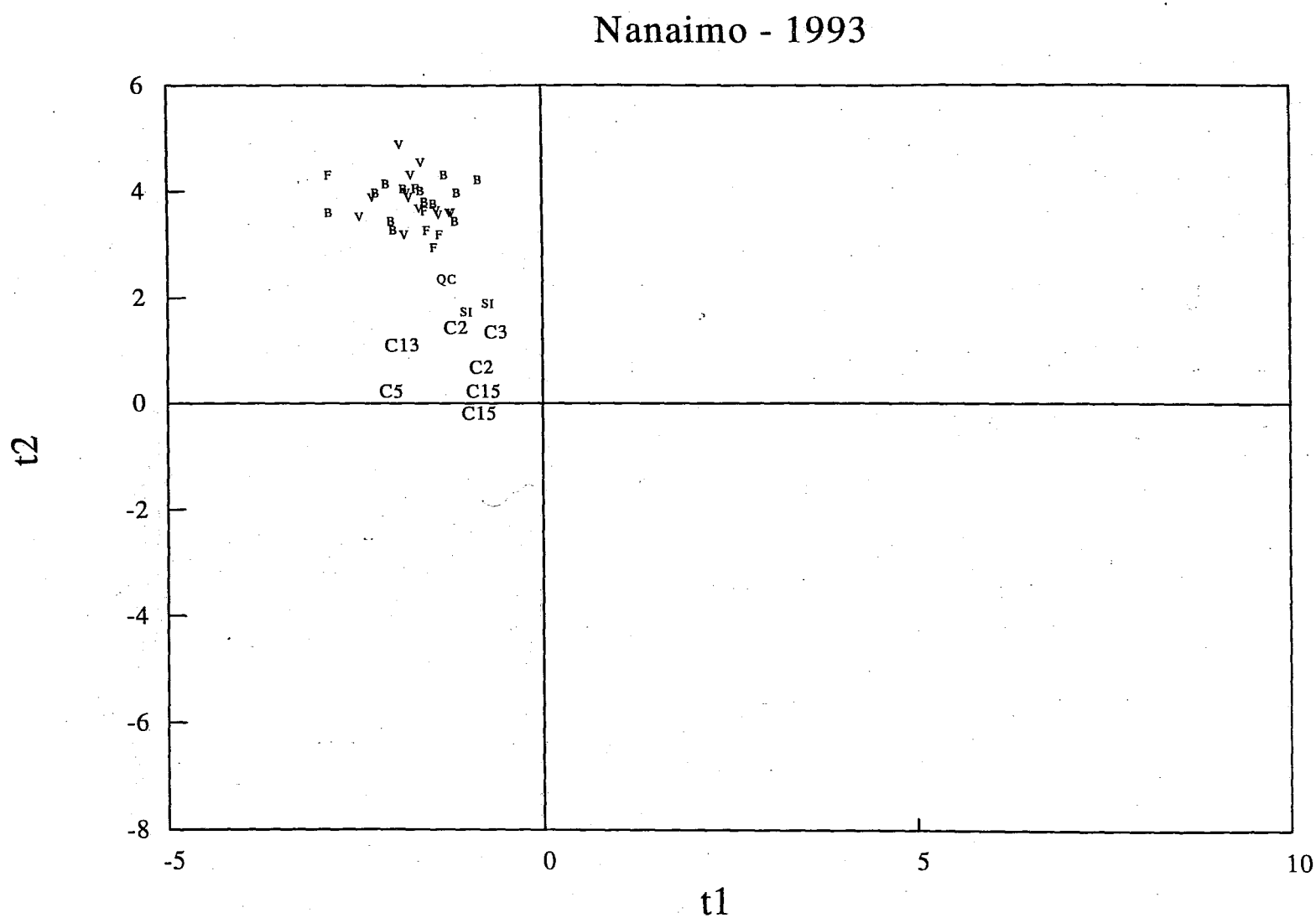


Figure 9-8. PCA projections for samples from Nanaimo in 1993.

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10. PORT ALBERNI

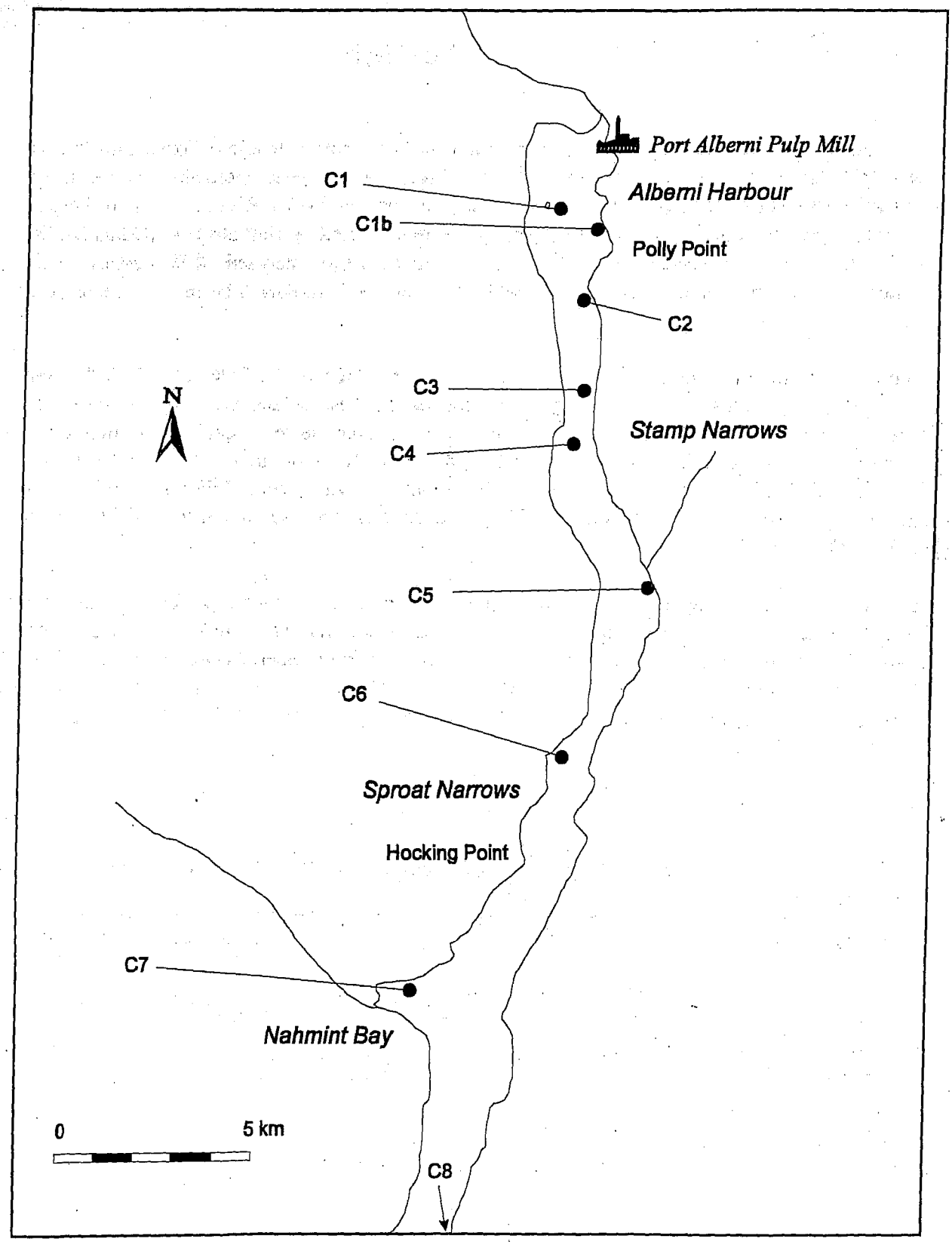
Crab samples were collected in Alberni Harbour and Alberni Inlet by Hatfield Consultants Ltd. in July 1990 (Map 10-1); one of these samples (C6) had too few congeners detectable to be retained in the PCA model (Table 2-2). Subsequent samples for the MacMillan Bloedel Limited Alberni Pulp and Paper Division mill monitoring program were collected by Hatfield Consultants Ltd. in March 1991, January 1992 and April 1993. Axys Analytical Ltd. analyzed all Port Alberni mill monitoring program samples from 1990 to 1993. All samples from Port Alberni were Dungeness crab.

Samples from Alberni Inlet have a higher proportion of the PnCDFs, HxCDFs and HpCDFs than most mill site samples, but a much lower proportion of these furans than any of the major harbours (Figure 10-1 and Figure 10-2; cf. Figure 3-3). Over the four years of the monitoring program the dibenzo-*p*-dioxins (with non-2,3,7,8 HxCDDs predominant) and the PnCDFs, HxCDFs and HpCDFs have persisted in the Alberni samples while the TCDFs have been largely removed. As a result congener totals and TEQs had decreased to one-half of the 1990 levels by 1993 (Table 10-1).

PCA projections for samples obtained throughout Alberni Inlet indicate a uniform composition that has remained distinct from the major B.C. harbours and that has changed little since 1990 (Figure 10-3 to Figure 10-6). In a few cases crab samples from Alberni Harbour (C1 and C1b) have varied from this composition. In 1991 and 1992 these samples had a PCA composition identical to the major harbours, but were indistinguishable from other Alberni Inlet samples in 1990 and 1993.

Table 10-1. Dungeness crab hepatopancreas congener total and TEQ averages for samples with similar PCA projections from Port Alberni.

Samples Averaged	**Congener Total**	**Calculated TEQ**
	pg/g wet weight	
Earliest Samples		
Port Alberni C1, C2, C4, C5, C6, C7, 1990 (n=6)	450	23
Most Recent Samples		
Port Alberni C1, C5, C6, C7, 1993 (n=4)	230	11



Map 10-1. Crab sampling locations near Port Alberni.

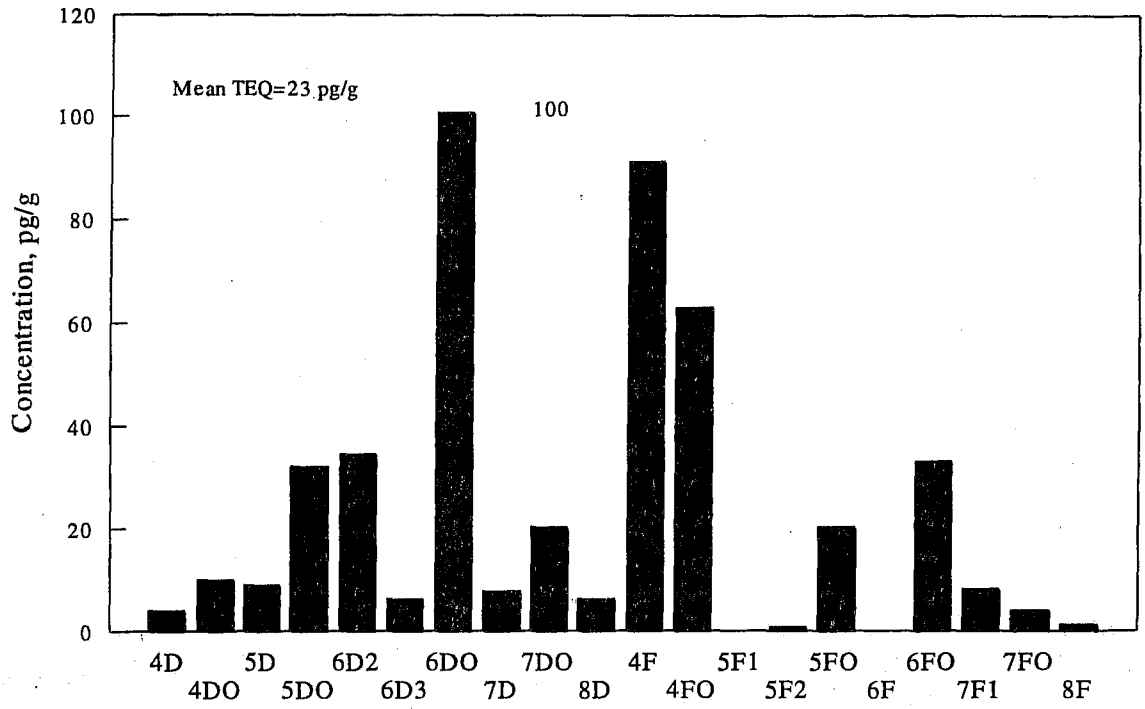


Figure 10-1. Port Alberni congener profiles for 1990 (n=6).

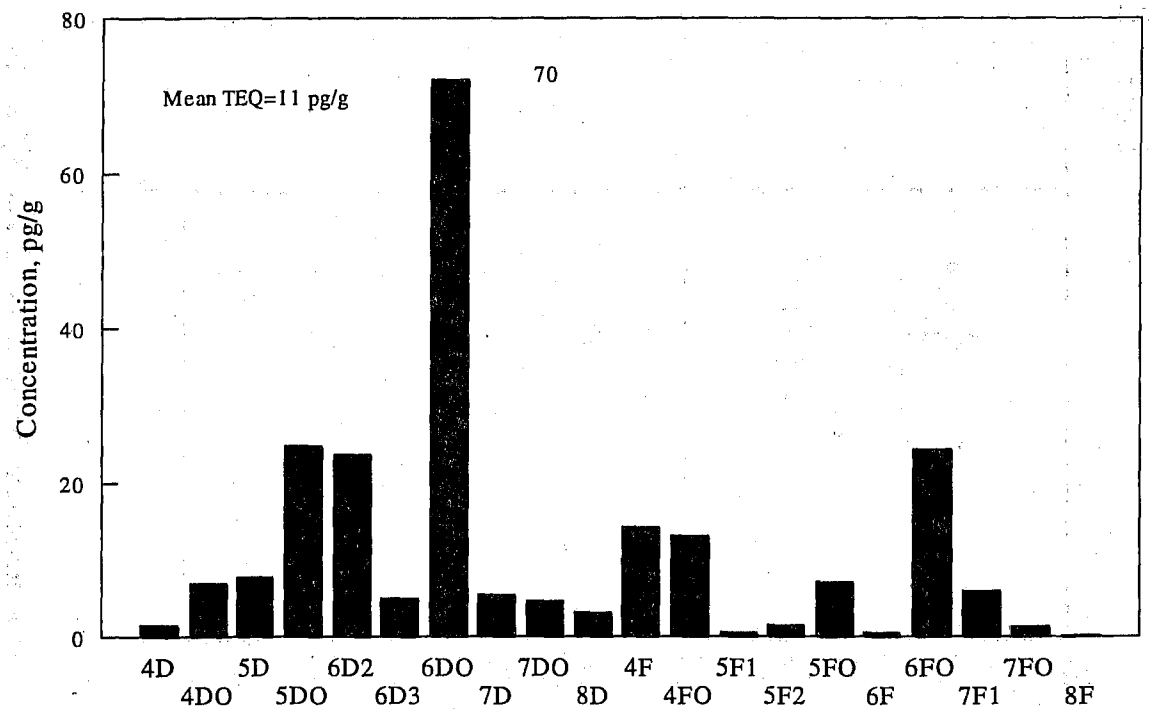


Figure 10-2. Port Alberni congener profiles for 1993 (n=4).

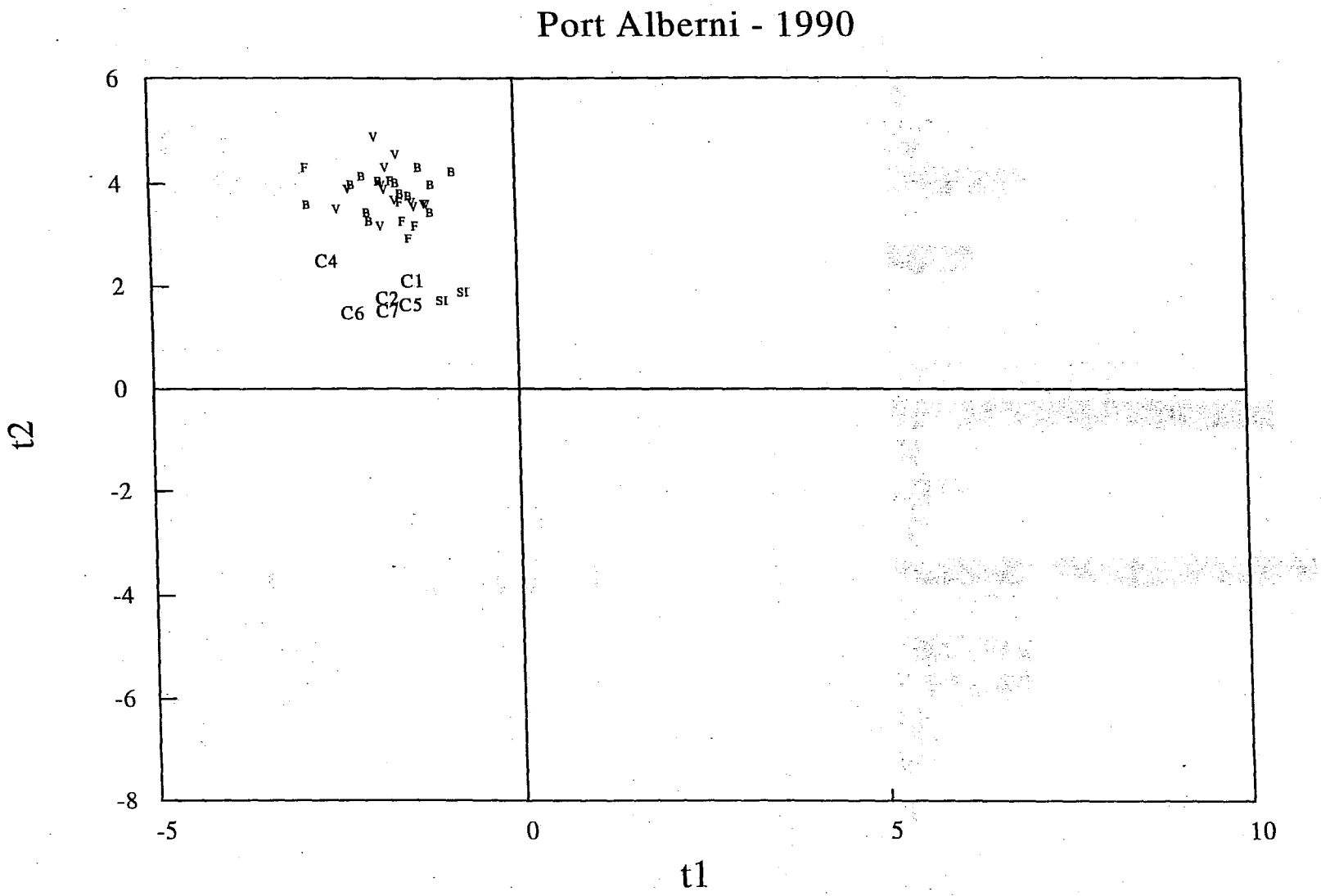


Figure 10-3. PCA projections for samples from Port Alberni in 1990.

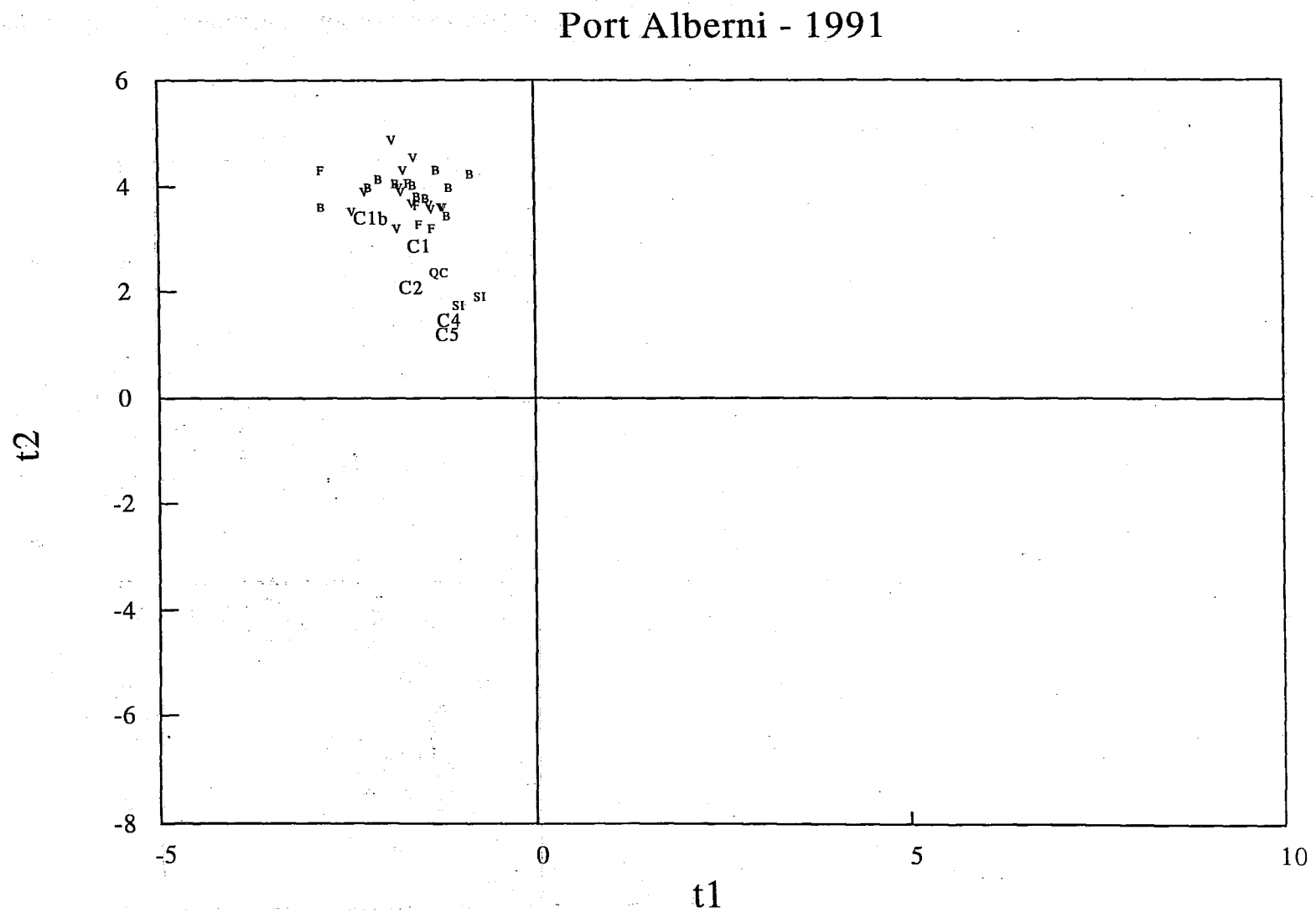


Figure 10-4. PCA projections for samples from Port Alberni in 1991.

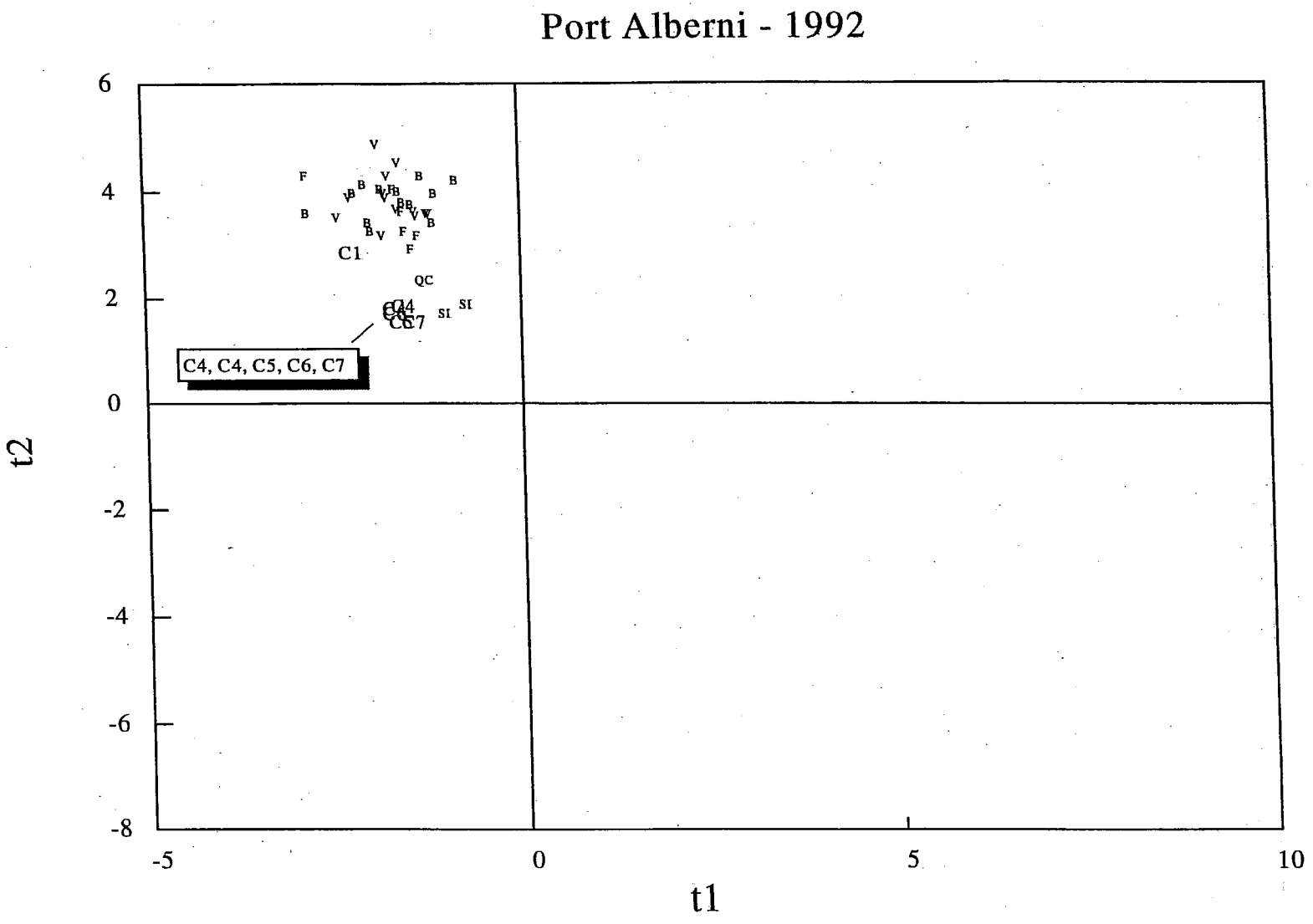


Figure 10-5. PCA projections for samples from Port Alberni in 1992.

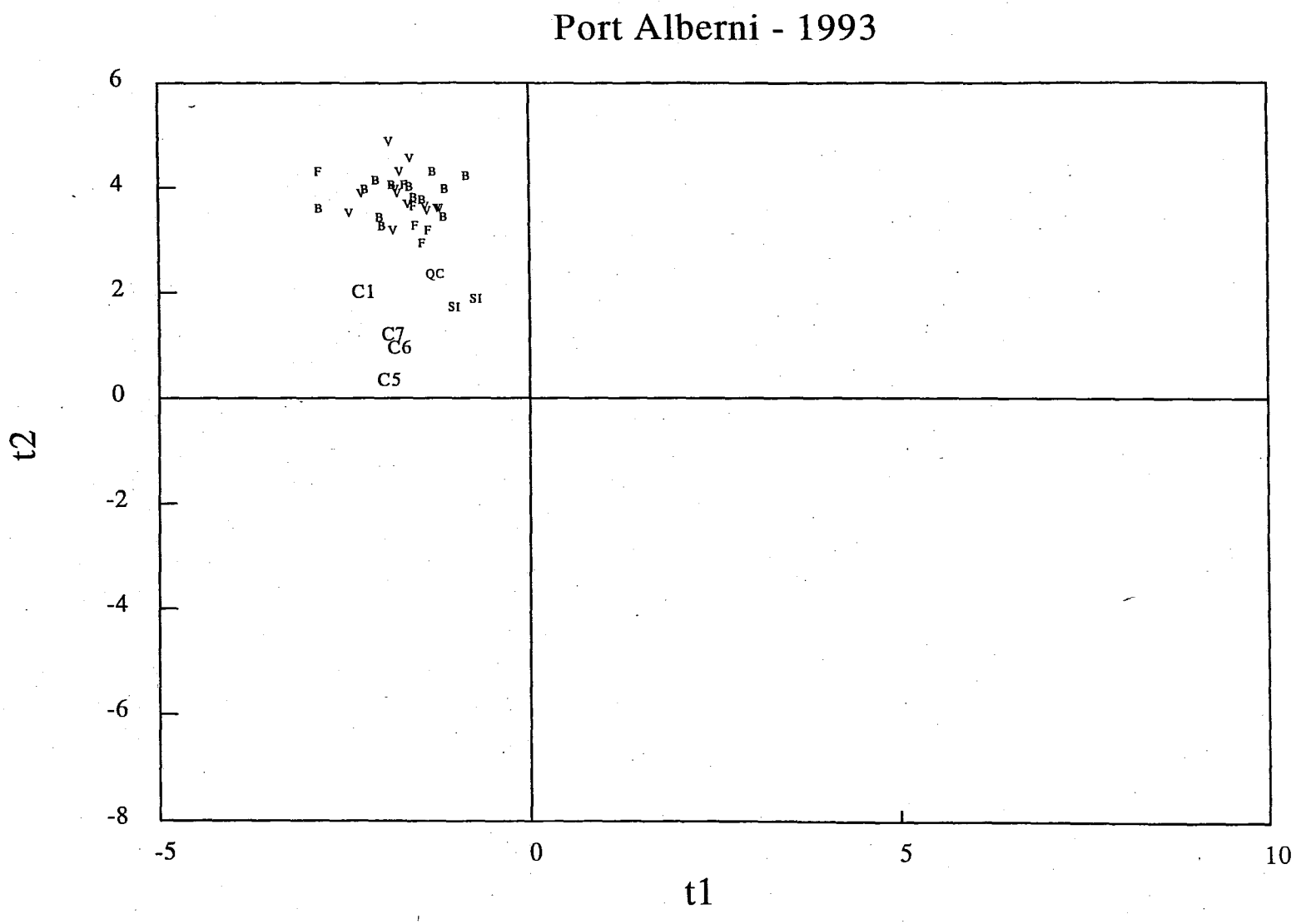


Figure 10-6. PCA projections for samples from Port Alberni in 1993.

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11. POWELL RIVER

Crab samples were collected in the Powell River area by Hatfield Consultants Ltd. in 1990, with five samples collected in the vicinity of the MacMillan Bloedel Limited Powell River Division Pulp and Paper mill in January and February and five supplementary samples collected further afield in September (Map 11-1). Three of these latter samples (C7, C8 and C11) had too few congeners detectable to be retained in the PCA model (Table 2-2). Subsequent mill monitoring program samples were collected by Hatfield Consultants Ltd. in March 1991, February 1992 and March 1993. Axys Analytical Ltd. analyzed all Powell River samples from 1990 to 1993. All PCA samples from the Powell River area were Dungeness crab except for samples of Box crab from Station C3 in 1990 and Red Rock crab from Stations C14 and C15 in 1993.

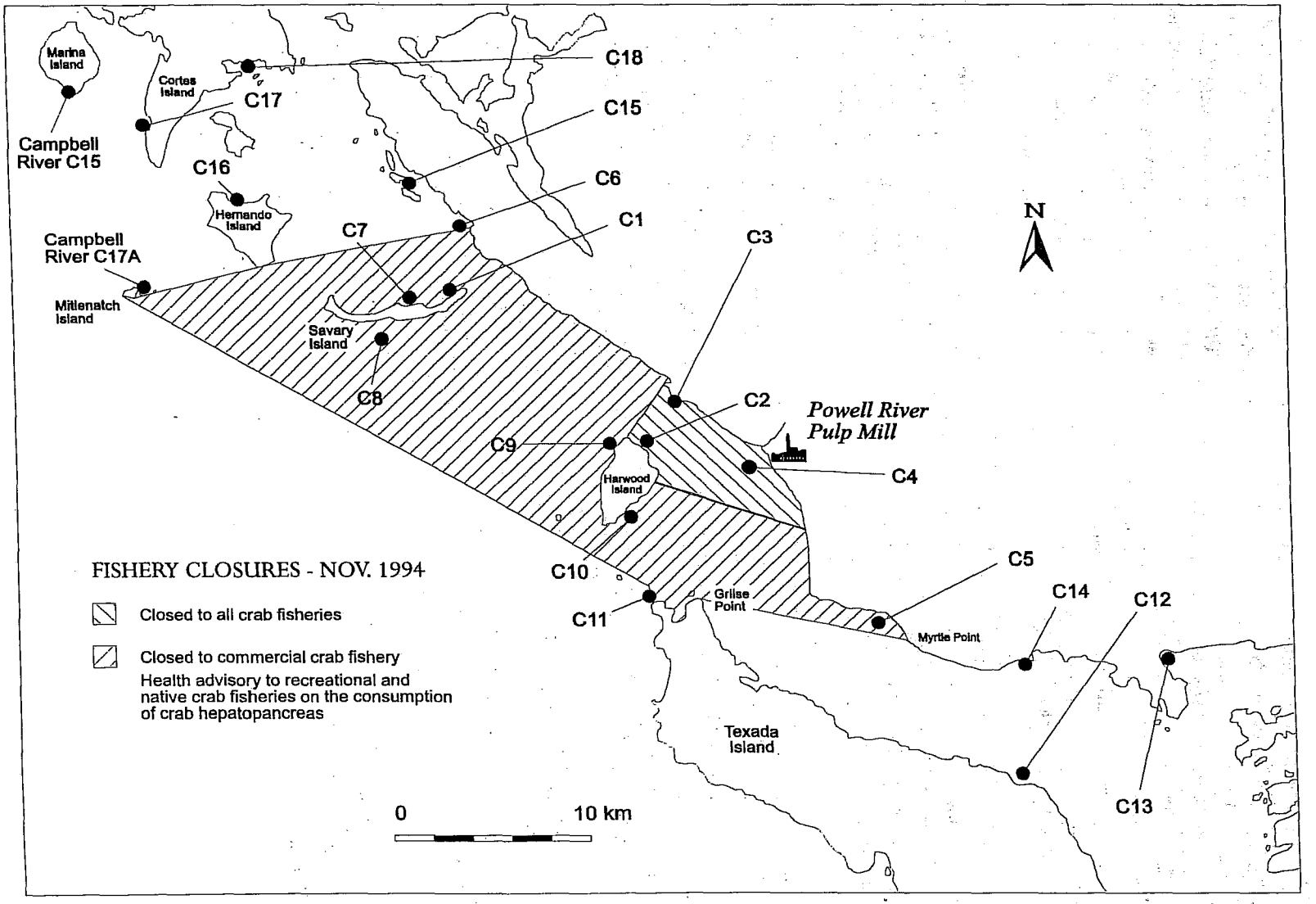
Between 1990 and 1993 the congener total and calculated TEQ decreased by approximately a factor of four for stations closest to the mill (Table 11-1). Congener profiles and PCA projections both indicated a decrease in the proportion of toxic congeners over this time (Figure 11-1 to Figure 11-6). The change in composition between years has been principally due to a decrease in the TCDFs, with an increase in the PnCDDs, HxCDDs, PnCDFs and HxCDFs.

From the PCA results it is evident that PCDD/Fs with a mill composition are present in crab samples ranging from Hernando and Cortes Islands (C16 to C18) in the northwest to Lang Bay (C14) and possibly Saltery Bay (C13) in the southeast (Figure 11-3 to Figure 11-6). PCA projections indicate that the congener composition has remained relatively consistent throughout the region in each year, indicating that the change in composition has proceeded uniformly over time. The Saltery Bay sample C13 from 1993 is very similar in PCA projection to samples that were collected between Howe Sound and Powell River in 1993 (Section 5).

The sample collected adjacent to the mill at Station C4 had PCA projections close to that of the harbours in 1993 (Section 4). Samples from this location were also shifted towards the harbour samples in 1991 and 1992. Because these samples were located closest to the mill, they very likely have been influenced by pentachlorophenol from contaminated wood chips.

Table 11-1. Dungeness crab hepatopancreas congener total and TEQ averages for samples with similar PCA projections from Powell River.

Samples Averaged	**Congener Total**	**Calculated TEQ**
	pg/g wet weight	
Earliest Samples		
Powell River C1, C2, C4, C5 and C9, 1990 (n=5)	620	57
Most Recent Samples		
Powell River C1 and C5, 1993 (n=2)	160	12



Map 11-1. Crab sampling locations near Powell River.

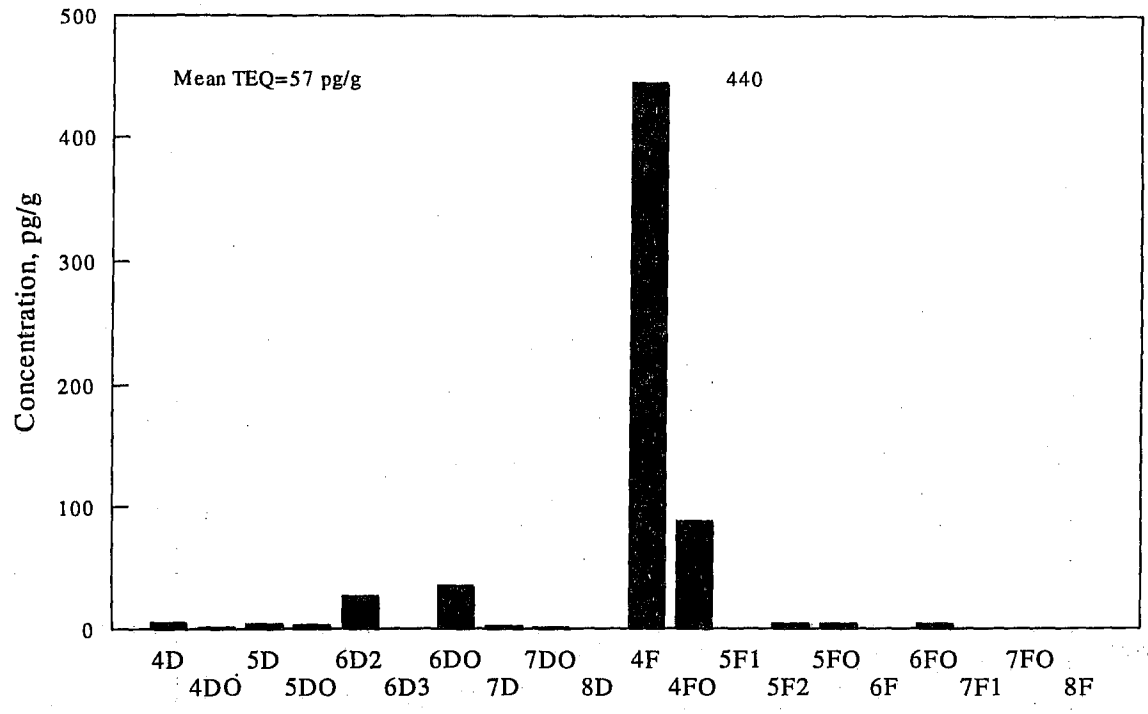


Figure 11-1. Powell River congener profiles for 1990 (n=5).

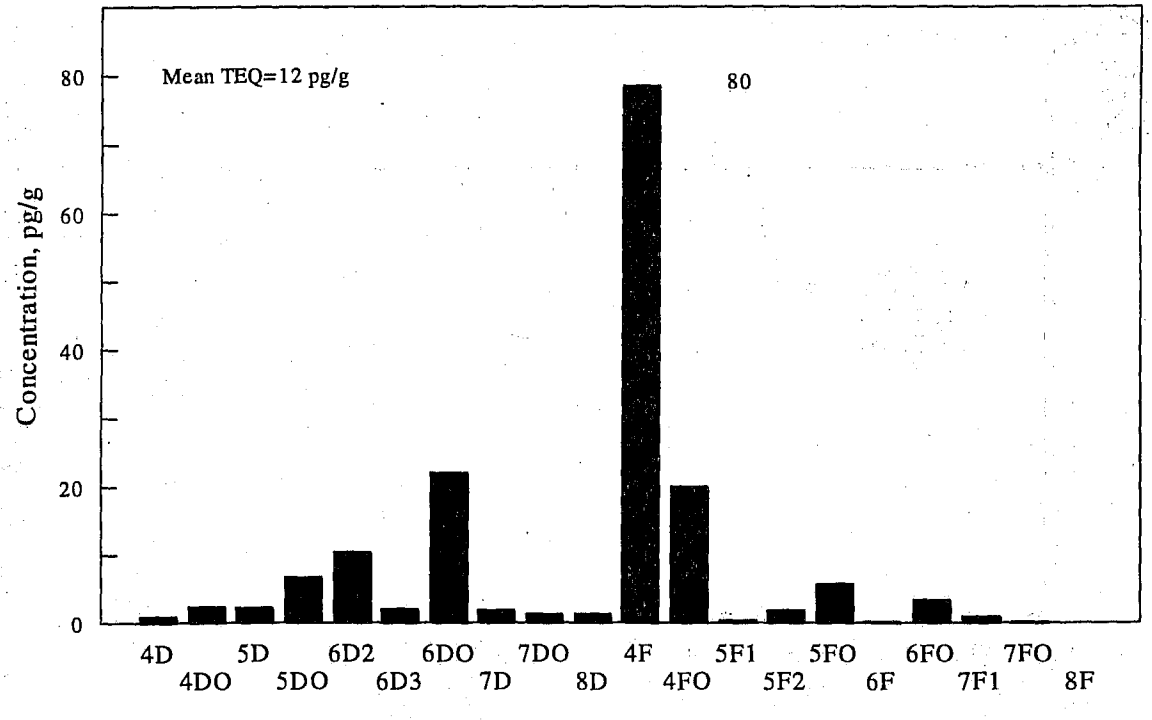


Figure 11-2. Powell River congener profiles for 1993 (n=2).

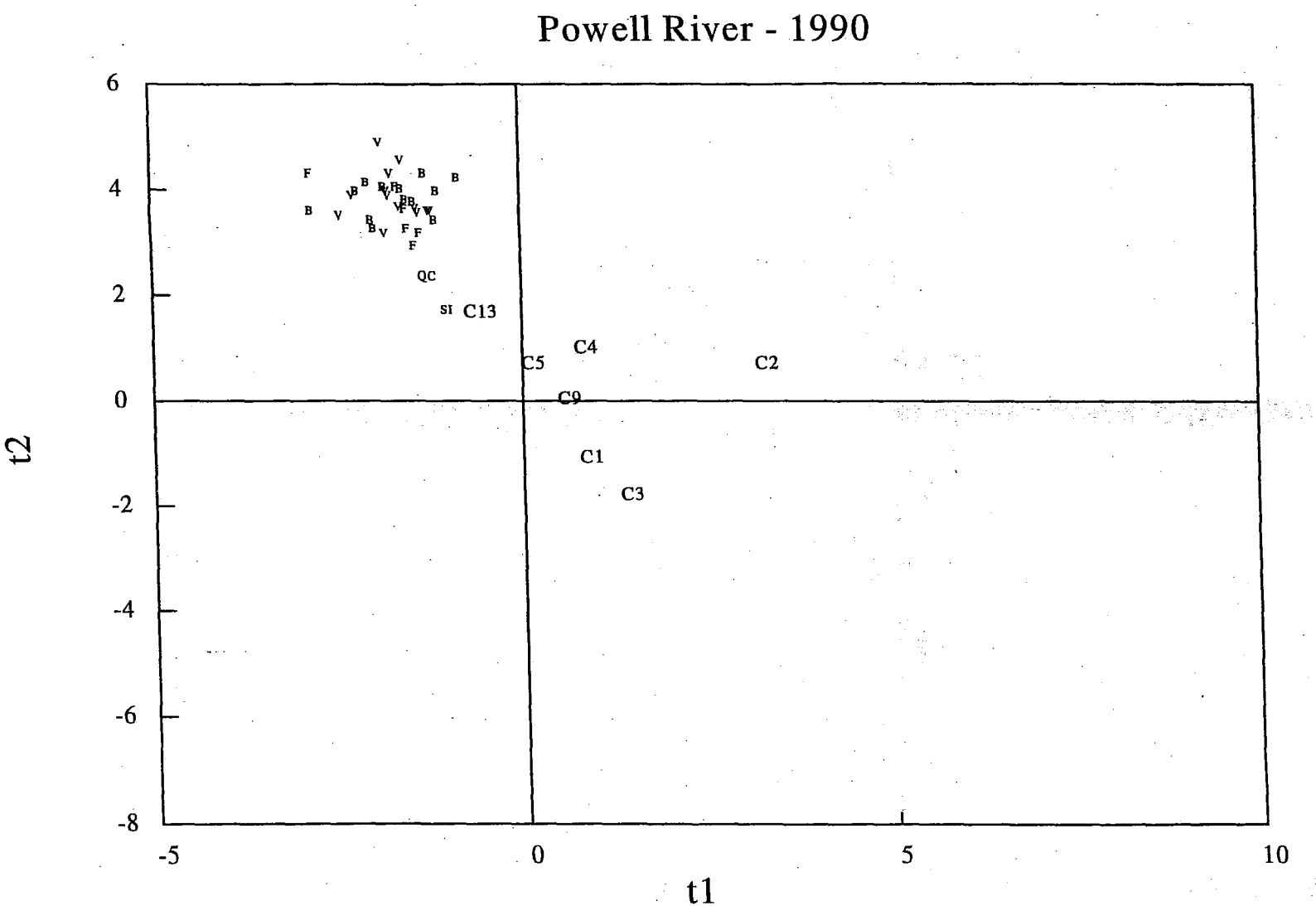


Figure 11-3. PCA projections for samples from Powell River in 1990.

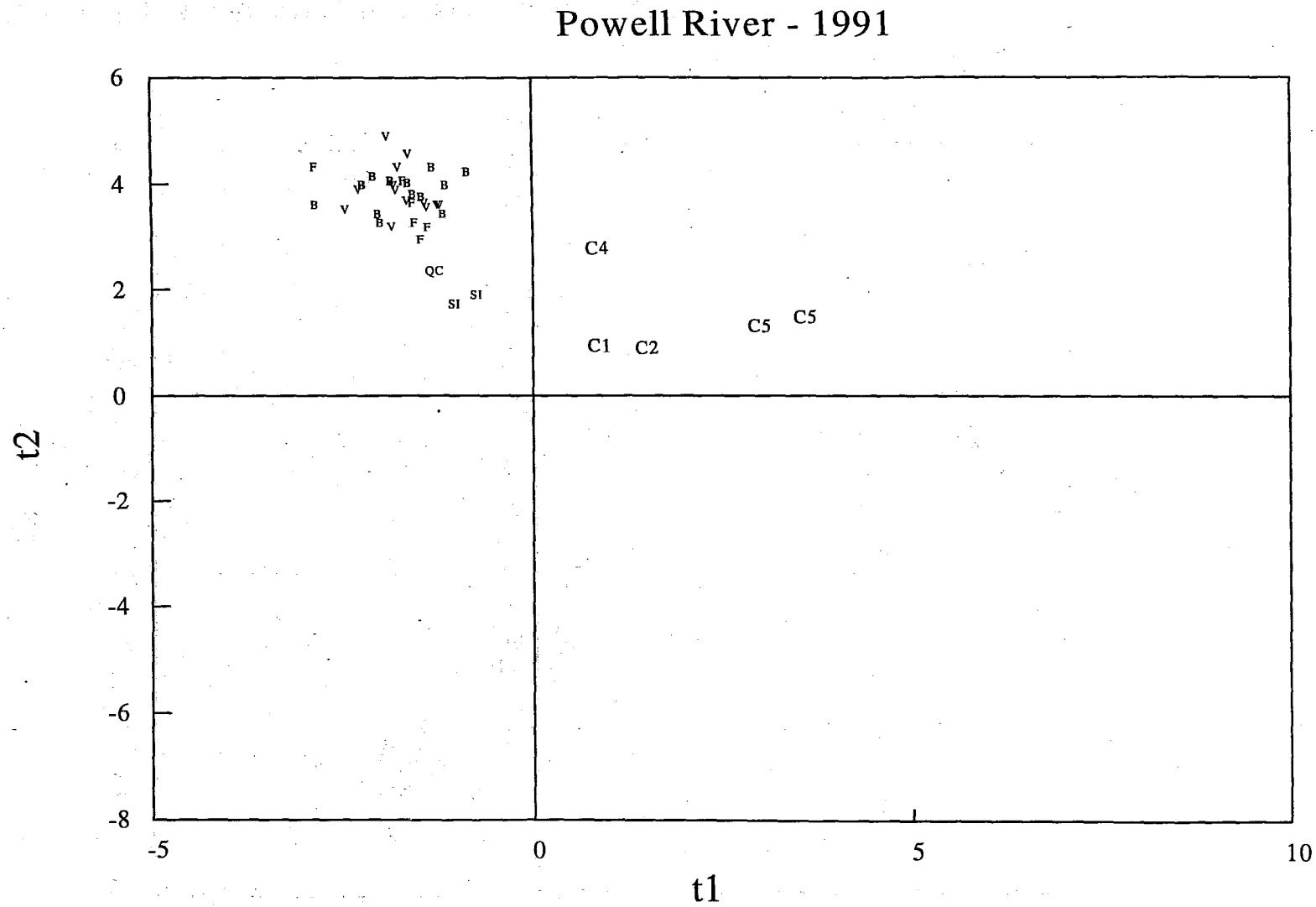


Figure 11-4. PCA projections for samples from Powell River in 1991.

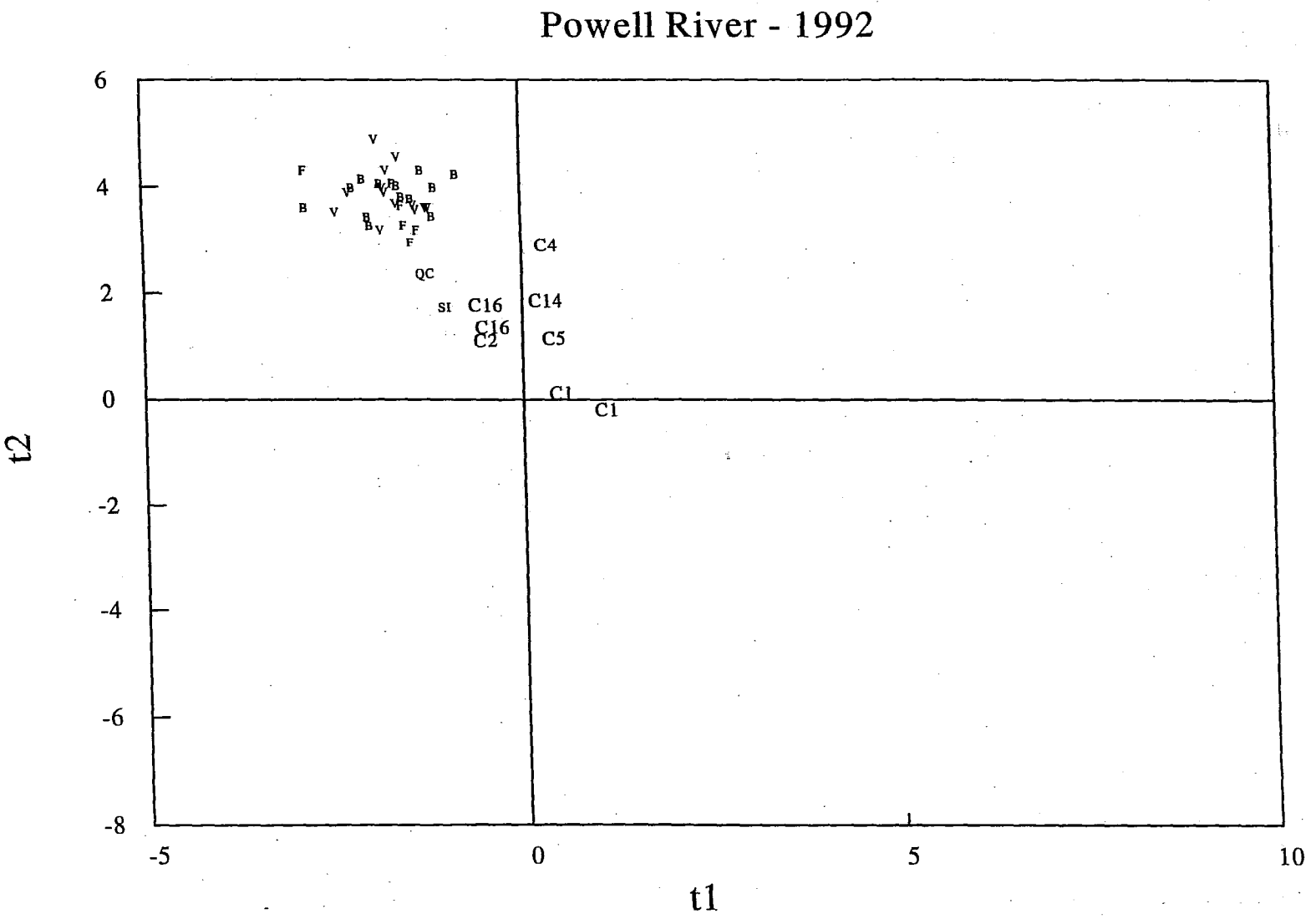


Figure 11-5. PCA projections for samples from Powell River in 1992.

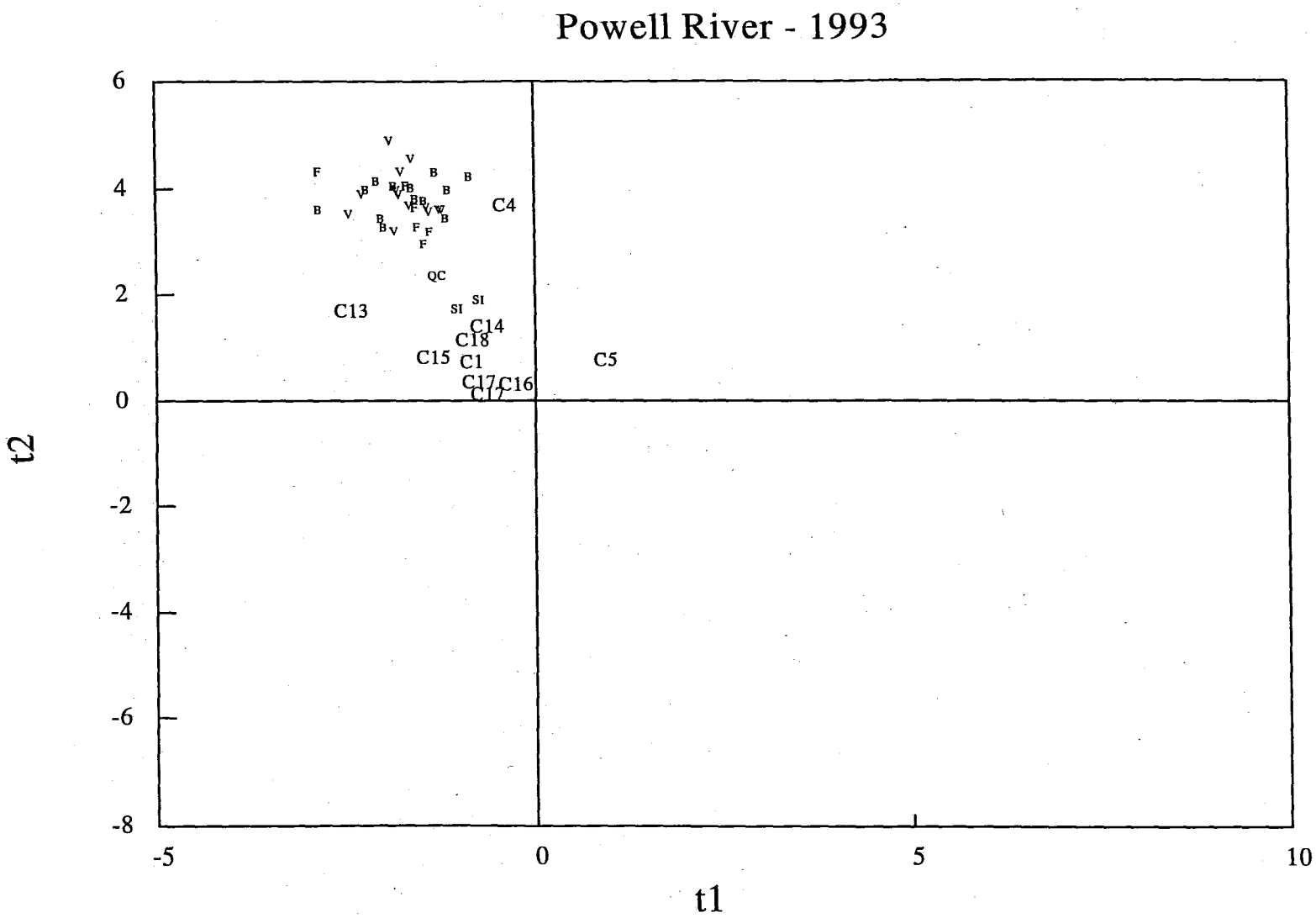


Figure 11-6. PCA projections for samples from Powell River in 1993.

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12. PRINCE RUPERT

Crab samples were collected near the Skeena Cellulose Inc. pulp mill in Prince Rupert by the Department of Fisheries and Oceans in the winter of 1987-1988 and analyzed by the Canadian Wildlife Service (Norstrom *et al.* 1988). Subsequent crab samples were collected in the vicinity of the mill by Hatfield Consultants Ltd. in January 1989, with more wide-ranging sampling instituted in June 1990 (Map 12-1). Mill monitoring program samples were then collected annually by Hatfield Consultants Ltd. in March or April from 1991 to 1993. One of the 1990 samples (SC1A) and three of the 1991 samples (SC4, SC5 and SC6) had too few congeners detectable to be retained in the PCA model (Table 2-2). Axys Analytical Ltd. analyzed all mill monitoring program samples from 1990 to 1993. In August 1993 an additional commercial fishery crab sample was collected in Chatham Sound by the Department of Fisheries and Oceans and analyzed by the Institute of Ocean Sciences. All samples were Dungeness crab.

Congener profiles and PCA projections indicate that only a small change in congener composition had occurred between 1987 and March 1991 for the crab samples collected closest to the mill (Figure 12-1; Figure 12-3 to Figure 12-5). These samples were principally from the narrow channels to the south and east of Kaien Island — Porpoise Harbour, Wainwright Basin and Morse Basin (samples SC1, SC2, SC3 and SC5; Map 12-1). The small change that did occur between years was principally due to an increase in the proportion of HxCDDs. Crab samples collected outside this enclosed area in 1990 and 1991 had a distinctly different composition that was closer to the pattern exhibited by the principal B.C. harbours. These samples included the 1990 samples SC4 and SC6 that were collected from islands to the west in Hecate Strait and the 1991 sample SC1A from the north side of Kaien Island in Prince Rupert Harbour (Figure 12-4 and Figure 12-5; Map 12-1).

By 1992 PCA projections for the crab samples collected close to the mill had shifted to a cluster just right of axis centre (Figure 12-6). This shift towards the upper left indicated a larger decrease in the proportion of TCDFs between 1991 and 1992 than had occurred up to 1991. Samples collected in Prince Rupert Harbour and to the north and west of Kaien Island in 1992 formed a separate cluster of samples that projected closer to samples from the major harbours.

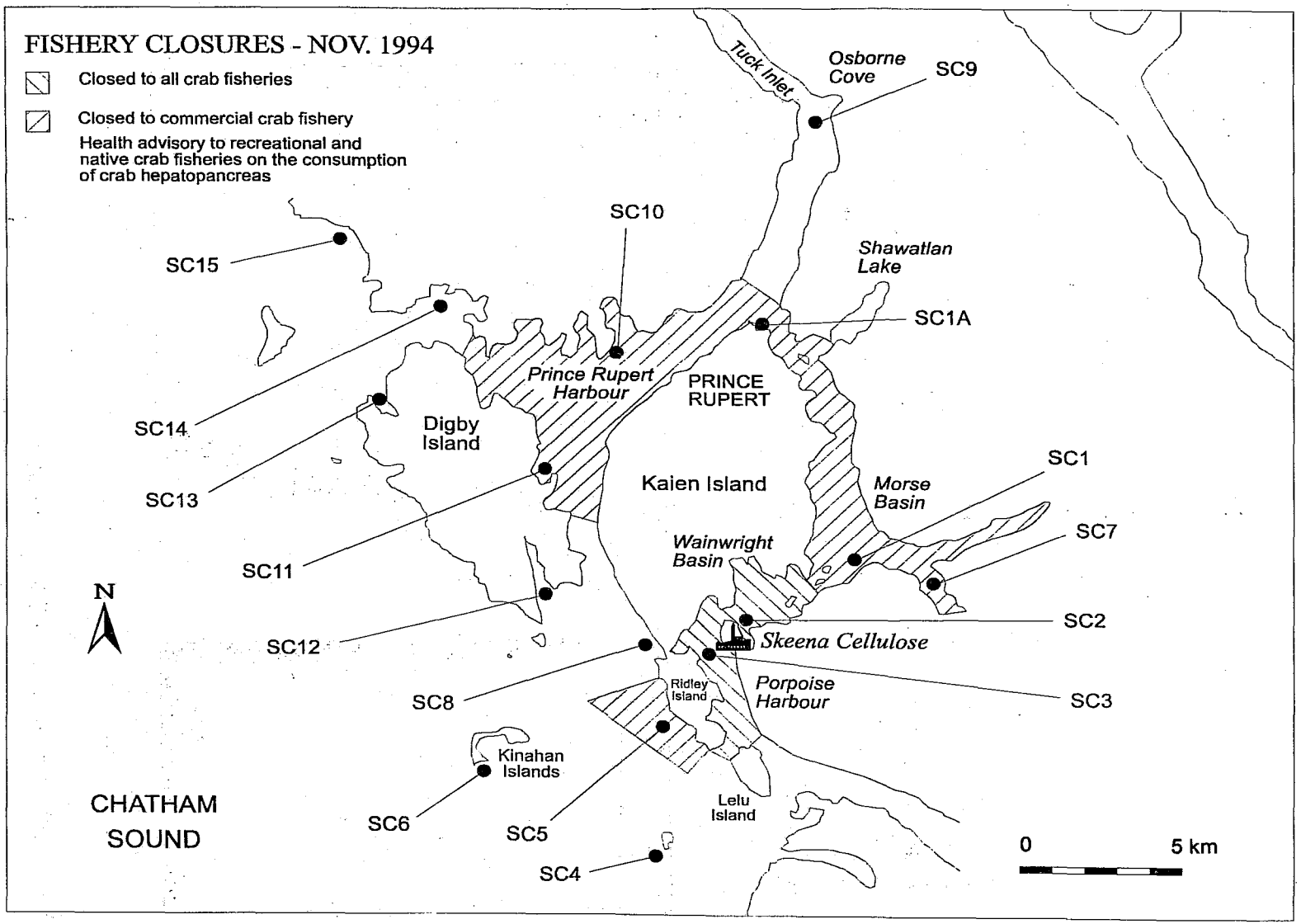
The progressive shift to the upper left in PCA projections, indicating a progressive increase in the HxCDD proportion, generally continued for most of the 1993 mill samples (Figure 12-7). One sample collected adjacent to the mill (SC2) had shifted in the opposite direction (toward the lower centre). This sample contained one of the highest proportions of non-2,3,7,8 HxCDDs in entire crab data set, as well as significant amounts of the TCDFs (not shown). By 1993 the HxCDD proportion was roughly twice the size of the individual TCDFs in the averaged mill samples (Figure 12-2). Samples from Prince Rupert Harbour in 1993 (SC1A, SC10, SC11) projected very close to samples from Victoria and Vancouver Harbours (Figure 12-7). Samples collected to the north and west again formed a distinct cluster between the harbour samples and the mill samples. The commercial fishery crab sample from Chatham Sound projected with this latter cluster.

As in other mill areas, PCA projections have remained relatively consistent at any given time in the channels adjacent to the mill; this indicates that the change in congener composition has proceeded uniformly over time. Between 1987 and 1993 the congener total and calculated TEQ have decreased by more than an order of magnitude for stations closest to the mill (Table 12-1).

Table 12-1. Dungeness crab hepatopancreas congener total and TEQ averages for samples with similar PCA projections from Prince Rupert.

Samples Averaged	**Congener Total**	**Calculated TEQ**
	pg/g wet weight	
Earliest Samples		
Weststar Pulp [a], 1987 (n=1)	17,600	1900
Prince Rupert SC1, SC3 and SC5, 1989 (n=3)	2900	260
Prince Rupert, 1987 (n=1) plus SC1, SC3 and SC5, 1989 (n=3)	6500	670
Most Recent Samples		
Prince Rupert SC1, SC2 and SC7, 1993 (n=4)	1000	53

[a] Since 1987 the name of the mill has changed from Weststar Pulp to Skeena Cellulose.



Map 12-1. Crab sampling locations near Prince Rupert.

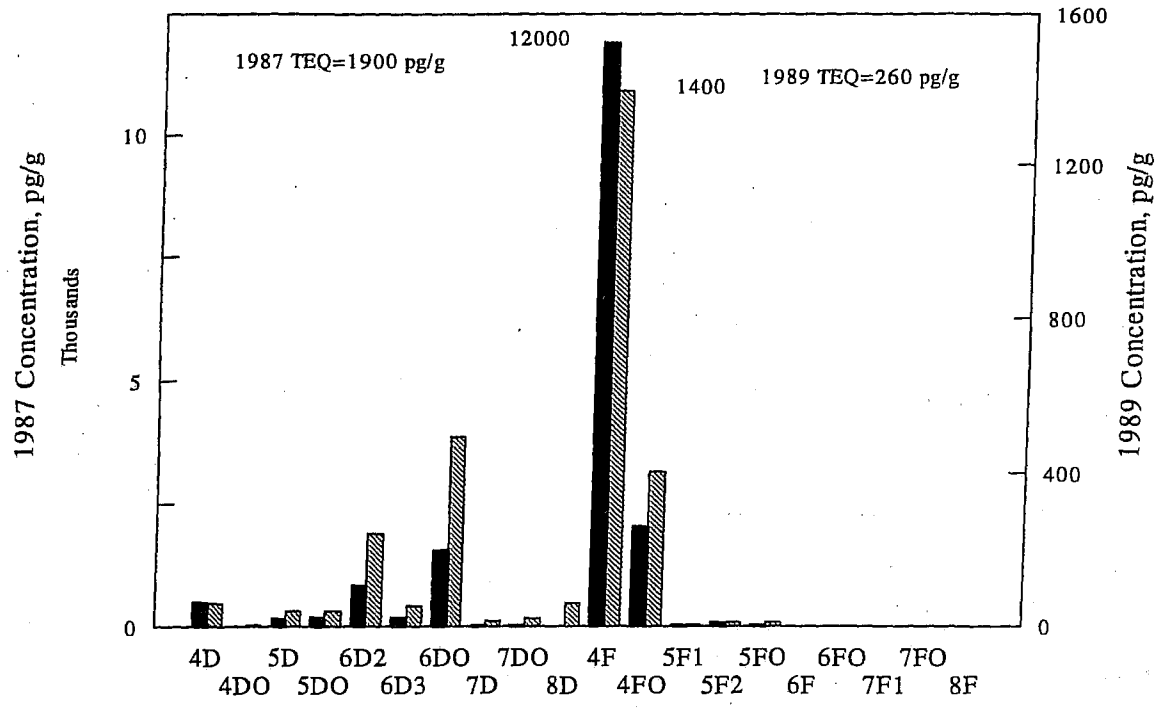


Figure 12-1. Prince Rupert congener profiles for 1987 (n=1) and 1989 (n=3).

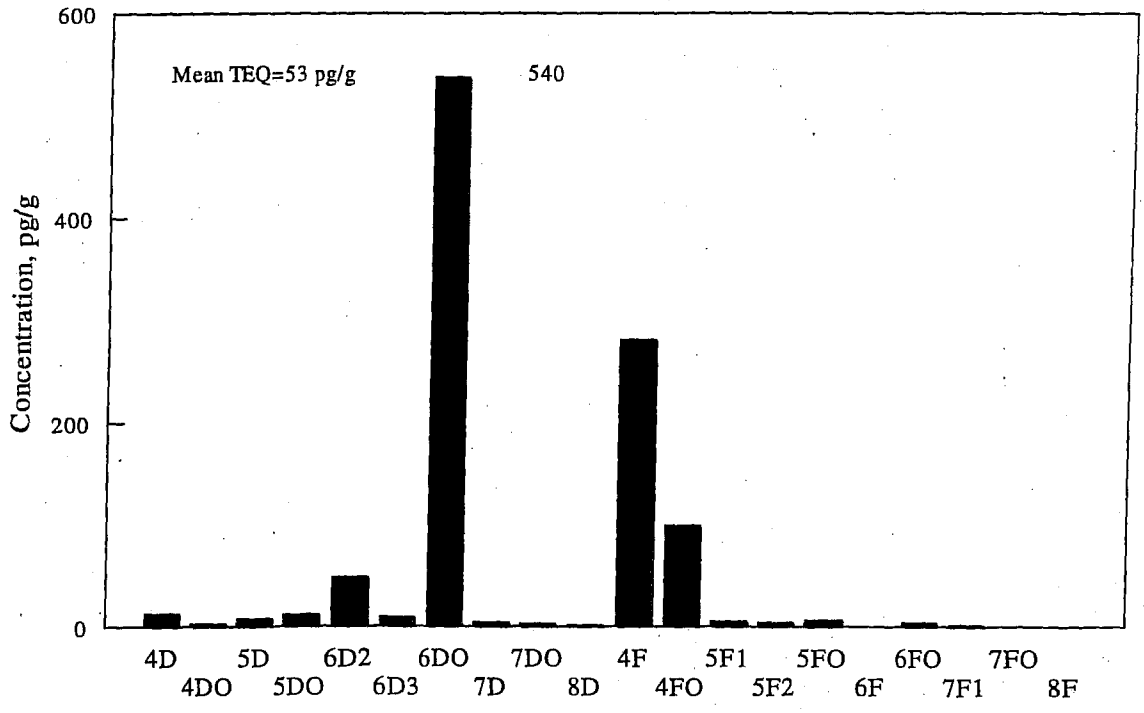


Figure 12-2. Prince Rupert congener profiles for 1993 (n=4).

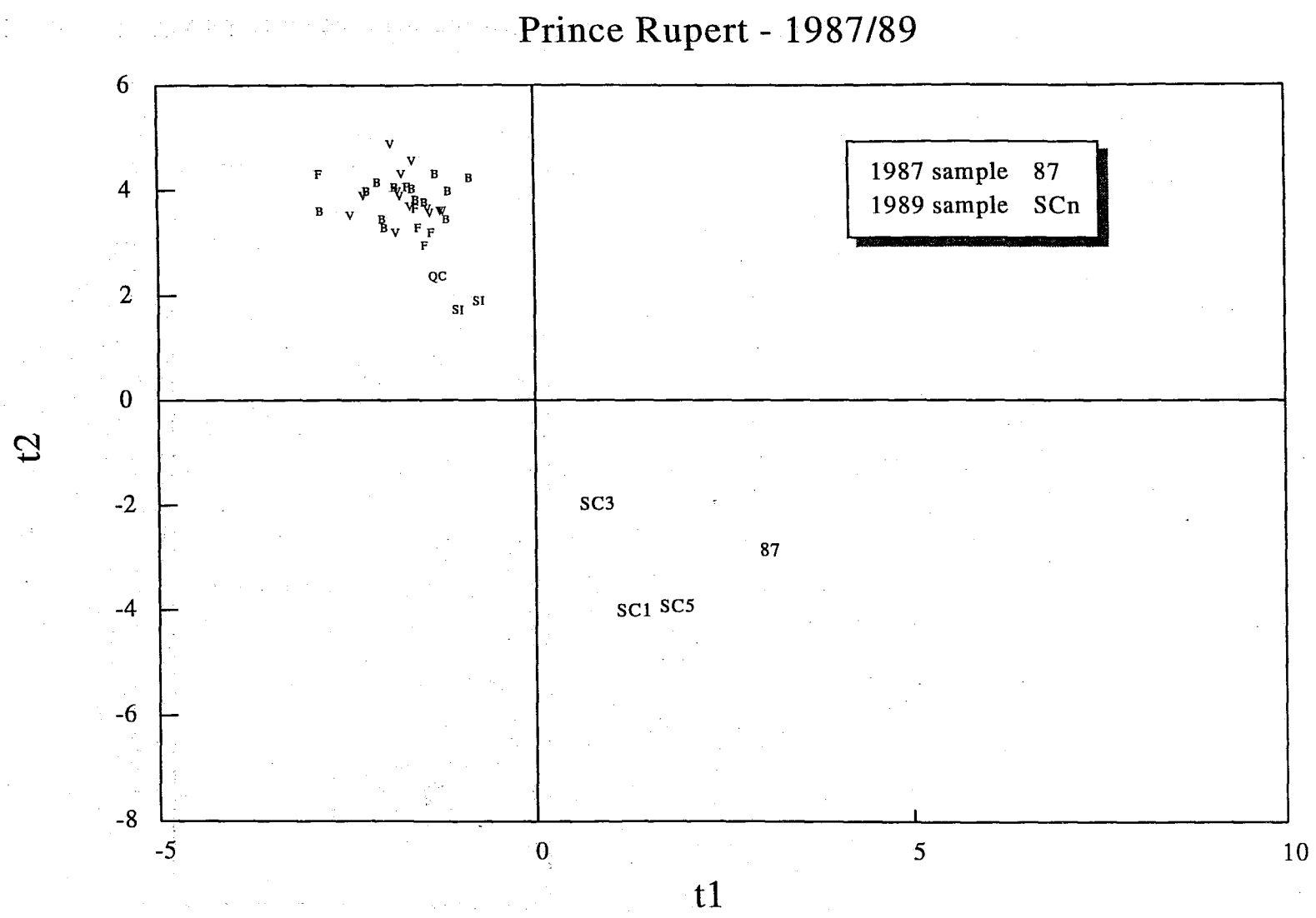


Figure 12-3. PCA projections for samples from Prince Rupert in 1987 and 1989.

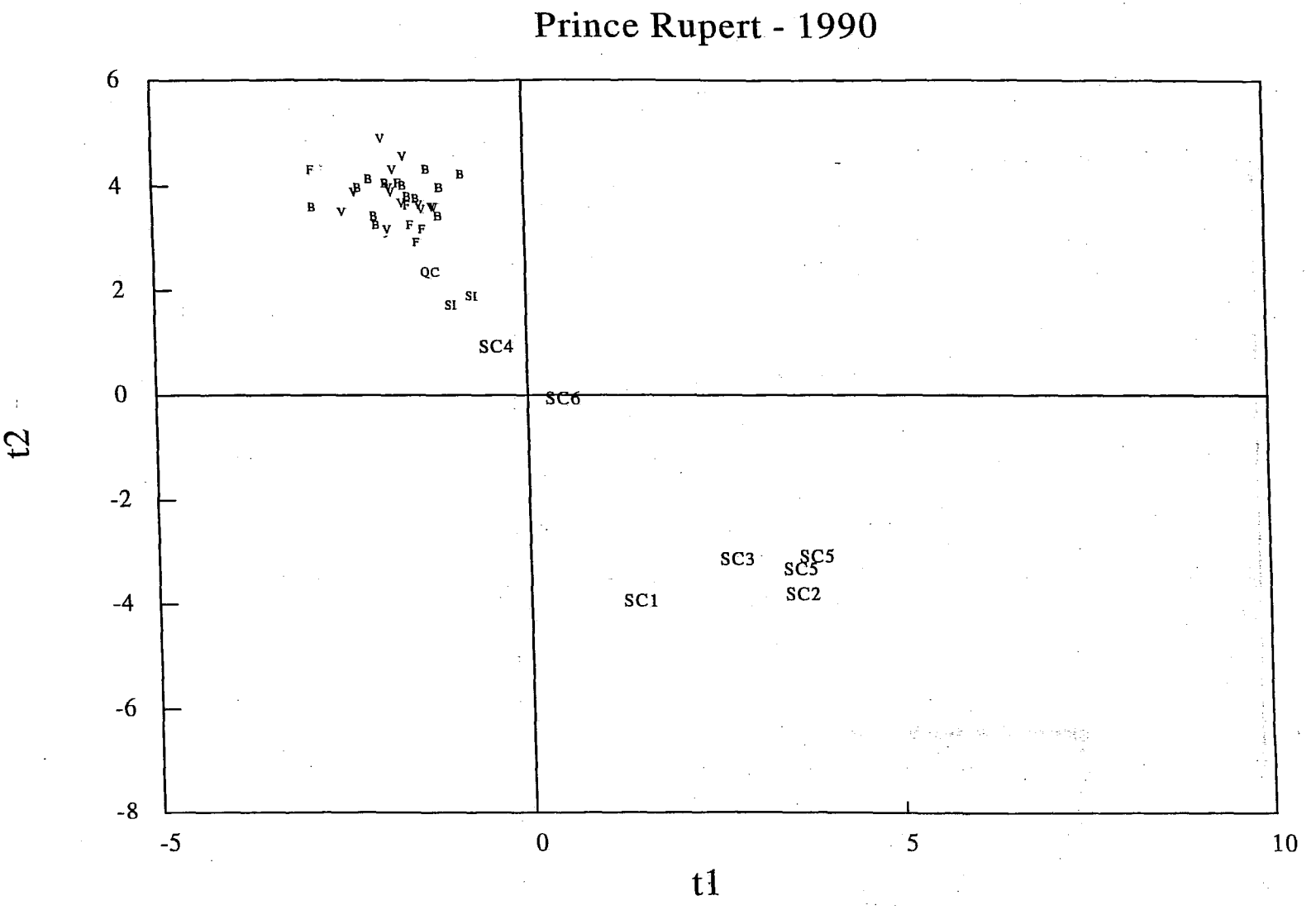


Figure 12-4. PCA projections for samples from Prince Rupert in 1990.

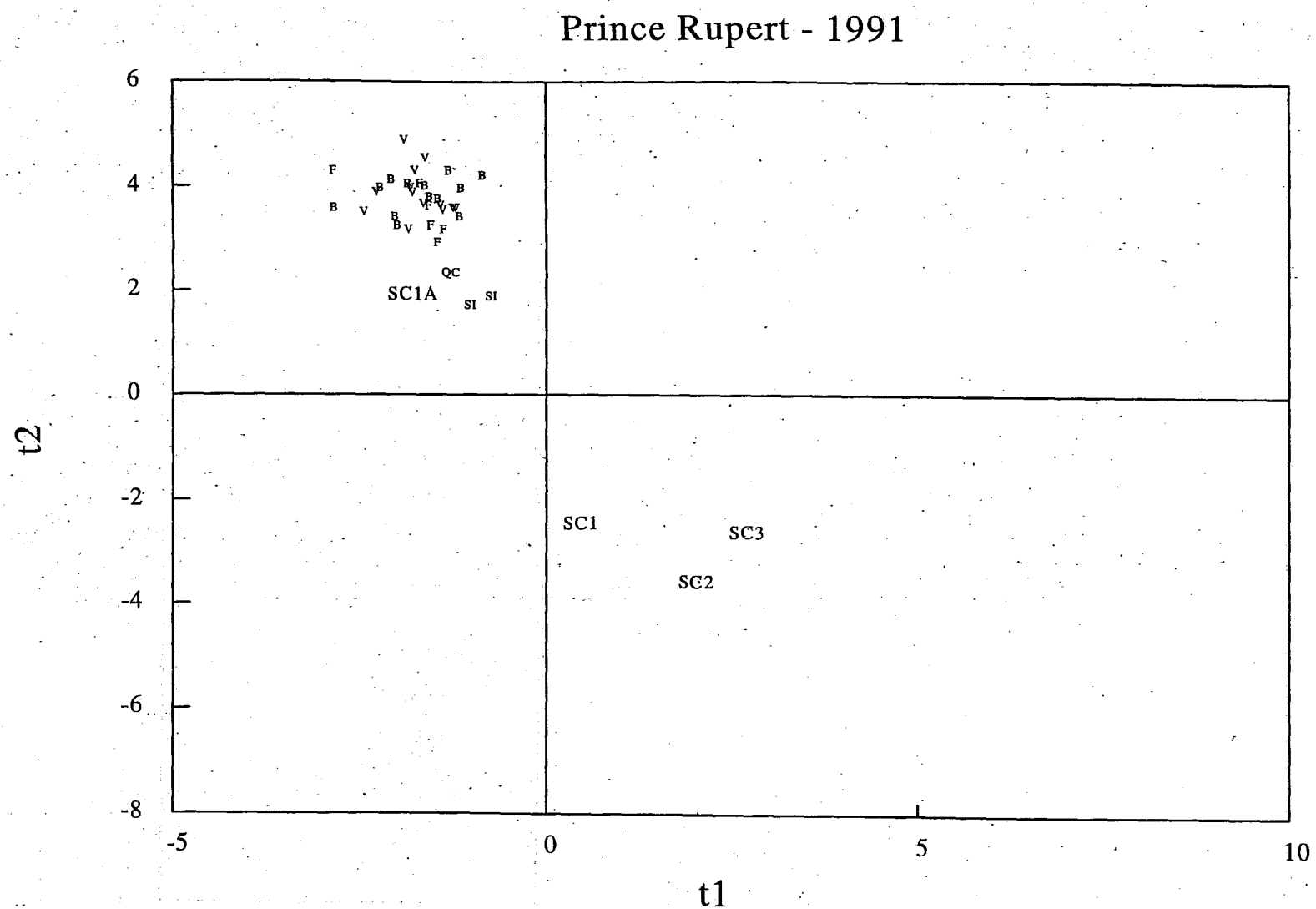


Figure 12-5. PCA projections for samples from Prince Rupert in 1991.

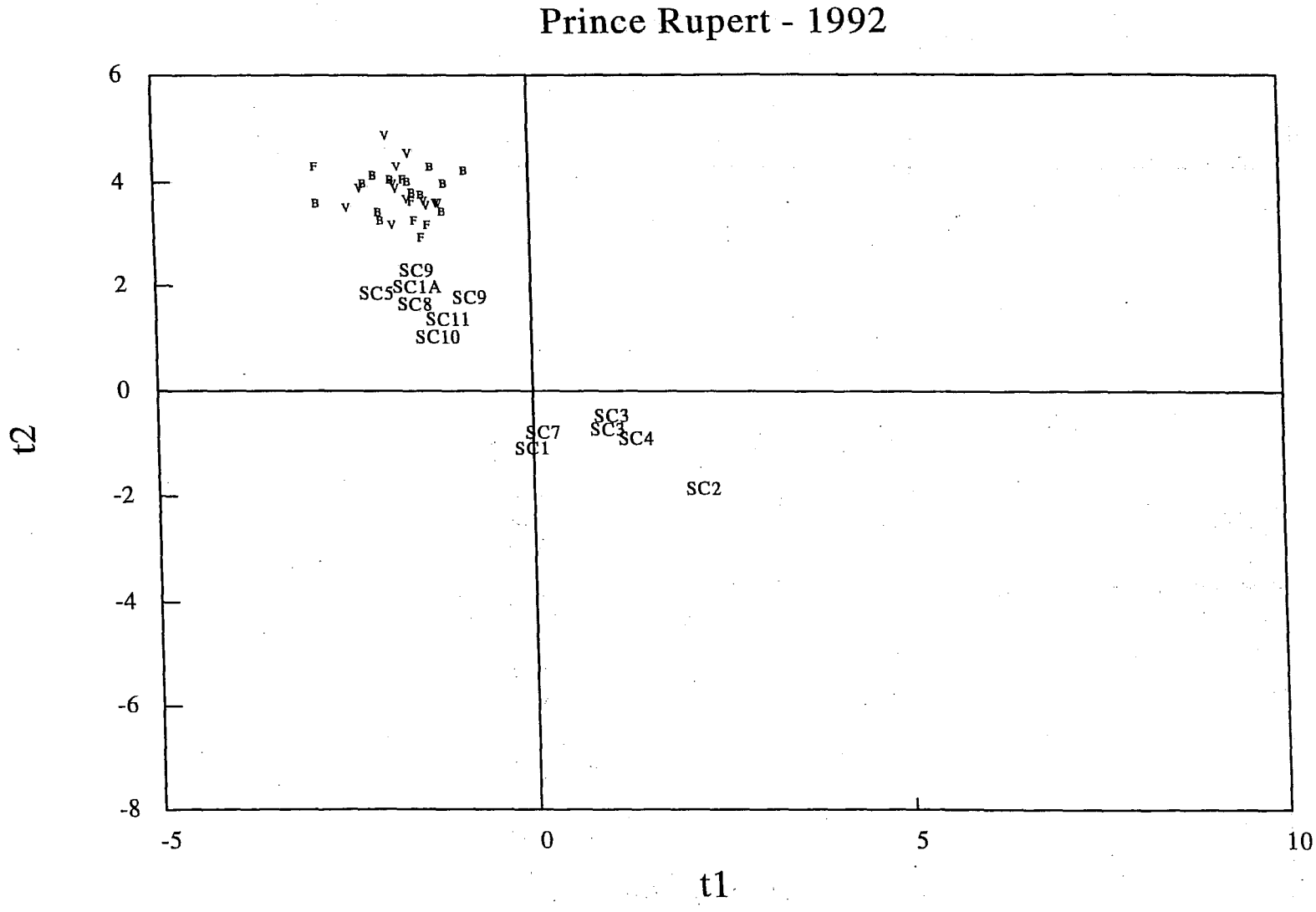


Figure 12-6. PCA projections for samples from Prince Rupert in 1992.

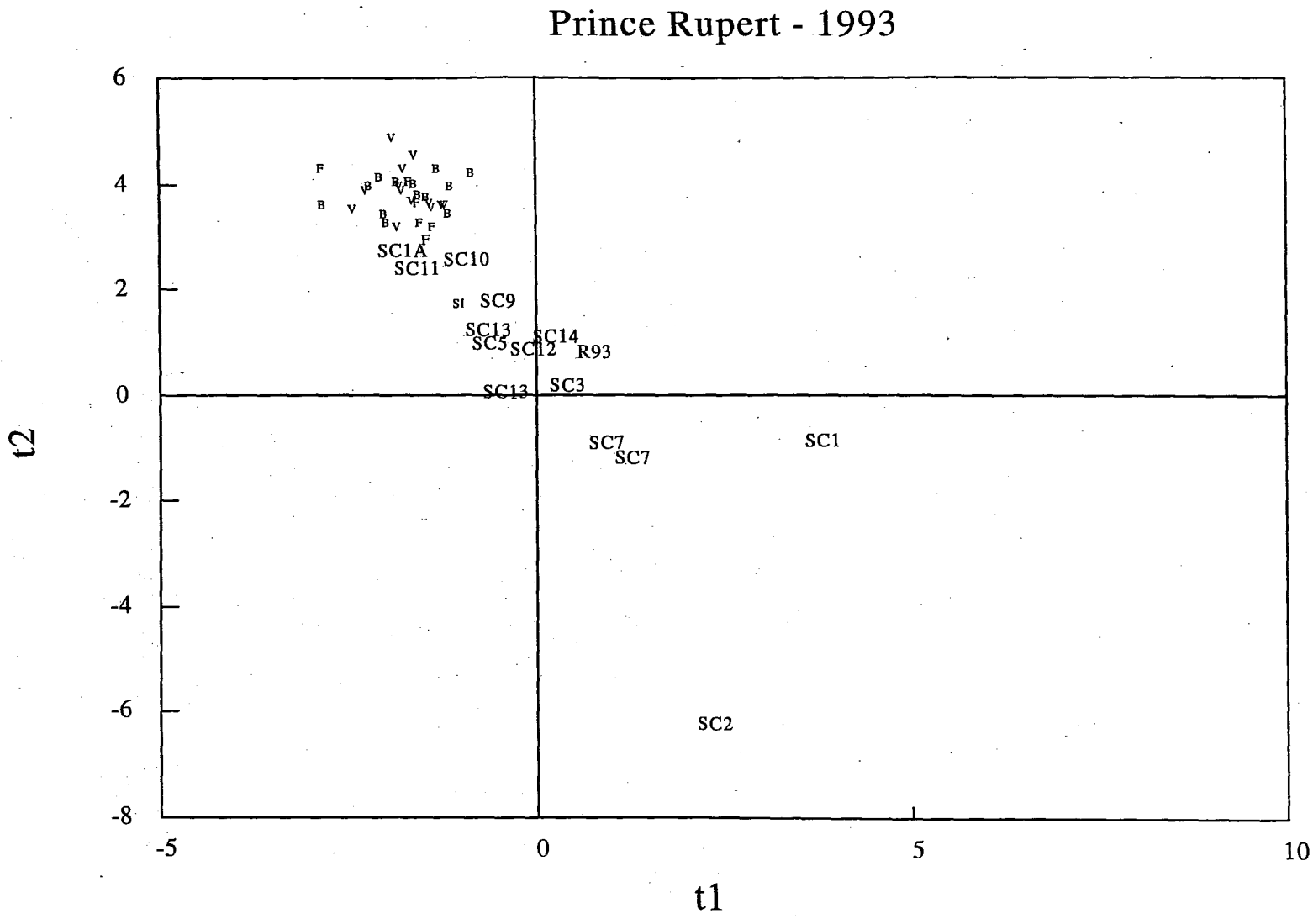


Figure 12-7. PCA projections for samples from Prince Rupert in 1993.

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13. KITIMAT

Crab samples were collected in Kitimat Arm by Norecol Ltd. in December 1989 and by the Department of Fisheries and Oceans in January 1991 and November 1992 (Map 13-1). A few locations to the south outside Kitimat Arm were also sampled in 1991. The 1989 sample from Gobeil Bay (S4) had too few congeners detectable to be retained in the PCA model (Table 2-2). Envirotest Ltd. of Edmonton Alberta analyzed the 1989 samples from Kitimat. The Burlington Ontario laboratory of the Department of Fisheries and Oceans analyzed the 1991 samples. Axys Analytical Ltd. analyzed the 1992 samples. All samples from Kitimat were Dungeness crab.

The Eurocan Pulp and Paper mill at Kitimat uses a Kraft pulping process, but it differs from the B.C. mills previously discussed because the mill does not use any chlorine bleaching. Accordingly, the TCDFs and PnCDFs that are characteristic of chlorine bleaching are almost completely absent in the Eurocan settling lagoon (Figure 13-1 and see following). Kitimat also has a major harbour, two other large industries — the Alcan aluminum smelter and the Methanex methanol production plant — and had a sawmill with a wood treatment facility from 1969 through 1981. Furthermore, pentachlorophenol treated lumber from another sawmill at nearby Terrace was apparently shipped from the docks at Kitimat through 1989 (Kelly, pers. commun., 1995). Hence high temperature processes at the aluminum smelter and pentachlorophenol used for wood treatment are both likely to have contributed additional PCDD/Fs to Kitimat fjord.

A number of samples are available to help characterize potential PCDD/F sources in the Kitimat area. Samples of sludge from five areas of the Eurocan Pulp and Paper Co. aeration lagoon were obtained by Norecol, Dames and Moore Inc. in 1993 and analyzed by Axys Analytical Ltd (congener profiles for each area were normalized before calculation of the average profile shown in Figure 13-1). A striking feature of the sludge samples is the almost complete absence of PCDFs. OCDD and the non-2,3,7,8 HxCDDs are major constituents in all sludge samples, but most of the PnCDDs in the averaged profile are due to one area of the lagoon. The non-2,3,7,8 HxCDDs also predominate in a sediment core collected by Environment Canada from Minette Bay in May 1990 (Figure 13-1). With the exception of OCDD, the averaged congener profiles for the lagoon and the sediment core are almost identical. Samples of roof dust, soil and sludge from the Alcan smelter site were collected by B.C. Environment in 1991 and also analyzed by Axys Analytical Ltd (Figure 13-2 and Figure 13-3). The HpCDDs, OCDD and the PCDFs that are the main constituents in the Alcan samples are only minor constituents in the sediment core.

Congener profiles for the 1989 crab samples from Kitimat Arm contain mainly HxCDDs with only minor amounts of the TCDFs; this pattern is consistent with polychlorinated phenol condensation, with little or no chlorine bleaching (Figure 13-4; cf. Section 3). The HxCDD, and to a lesser extent PnCDD, profile in crabs is mirrored in the sediment core from Minette Bay (Figure 13-1).

Between 1989 and 1992 the absolute concentrations of PnCDD and the HxCDDs (i.e. the congeners most characteristic of polychlorophenol condensation) and the TEQ have decreased by

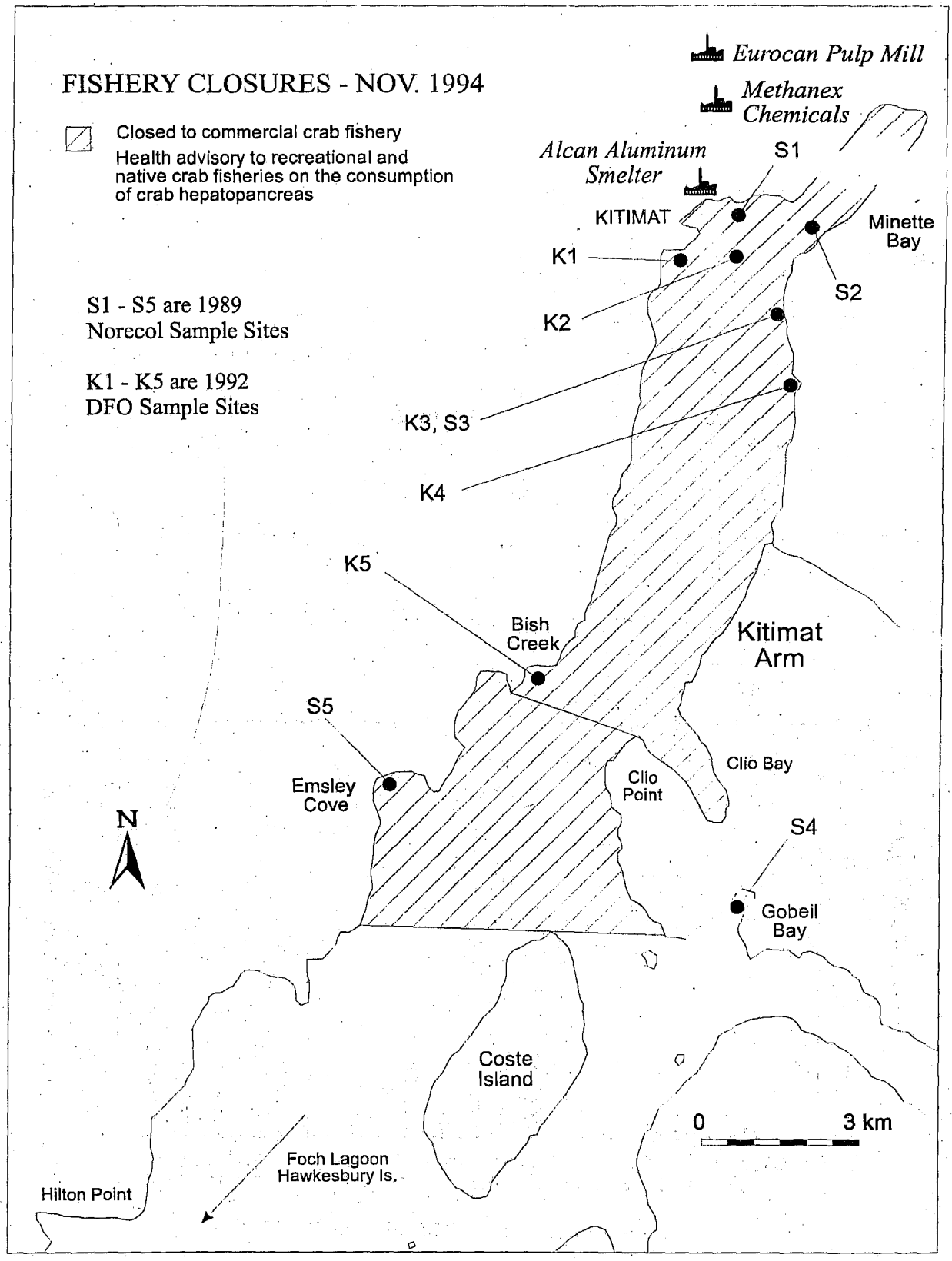
nearly an order of magnitude in Kitimat crab samples (Figure 13-4, Figure 13-5 and Figure 13-6; Table 13-1). Over this time period congener profiles have also shown an increase in the proportion of the non-2,3,7,8 substituted HpCDDs, as well as the TCDFs, PnCDFs and HxCDFs. These congeners were present in varying proportions in samples of soil and sludge and in some samples of roof dust collected on the Alcan aluminum smelter site in 1991 (Figure 13-2 and Figure 13-3). Again the match is not exact; the hepta- and octachlorinated congeners in particular were present in major amounts in the smelter samples, but are apparently not available to the crabs. The presence of the non-2,3,7,8 substituted congeners at the Alcan smelter site and in Kitimat Arm may not be due to recent contamination. It may be that these congeners were present in low levels in the 1989 crab samples, and are only becoming apparent as analytical detection limits improve.

The absence of chlorine bleaching and the presence of a novel suite of non-2,3,7,8 substituted congeners produces PCA projections for the Kitimat crab samples that are different from other regions of the B.C. coast (Figure 13-7 to Figure 13-9). Samples form a tight cluster on the PCA plot with only one sample from Bish creek in 1992 (sample 196) outside the cluster (Figure 13-9). While the overall PCA projections have changed little from 1989 to 1992, projections have generally shifted to the upper left as the HxCDD proportion has decreased (Figure 13-8 and Figure 13-9).

From the PCA projections of the 1991 samples it is evident that PCDD/Fs with the Kitimat Estuary composition are present in Gobeil Bay on Kildlala Arm (Map 13-1) and in Douglas and Sue channels at least as far as Foch Lagoon and Hawkesbury Island (Figure 13-8).

Table 13-1. Dungeness crab hepatopancreas congener total and TEQ averages for samples with similar PCA projections from Kitimat.

Samples Averaged	**Congener Total**	**Calculated TEQ**
	pg/g wet weight	
Earliest Samples		
Kitimat EC1, EC2, EC3 and EC5, 1989 (n=8)	570	68
Kitimat 1991, (n=7)	130	12
Most Recent Samples		
Kitimat 1992, (n=41)	110	7.2



Map 13-1. Crab sampling locations near Kitimat.

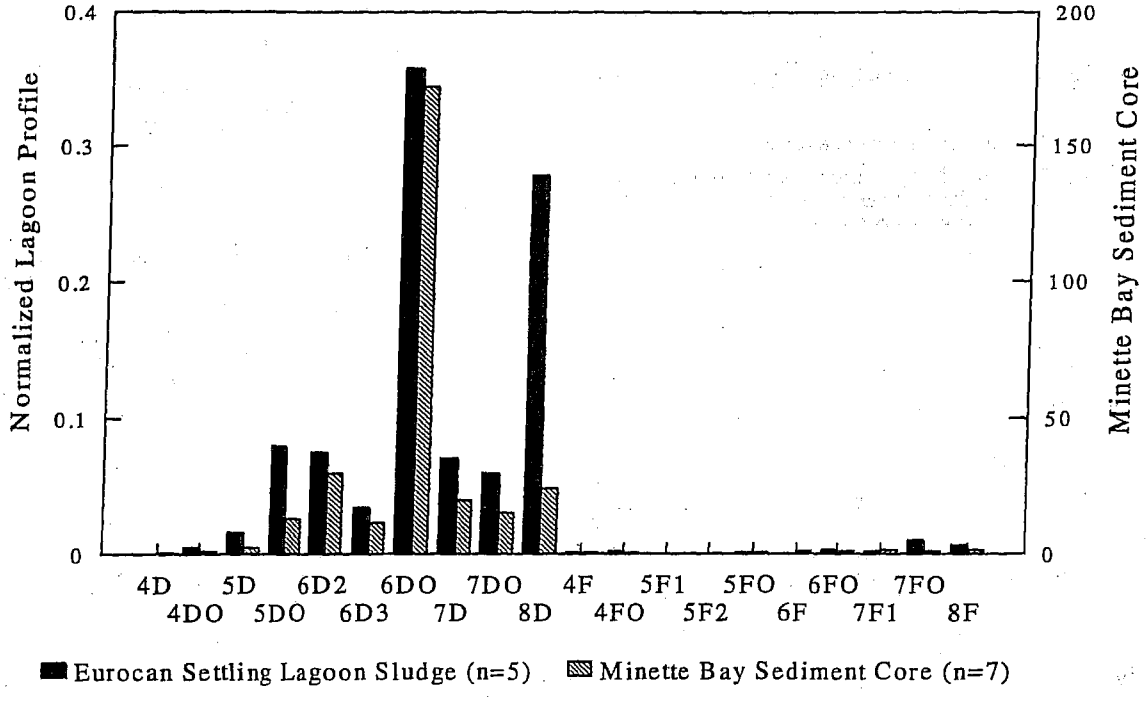


Figure 13-1. Congener profiles for Eurocan pulp mill sludge and Minette Bay sediment.

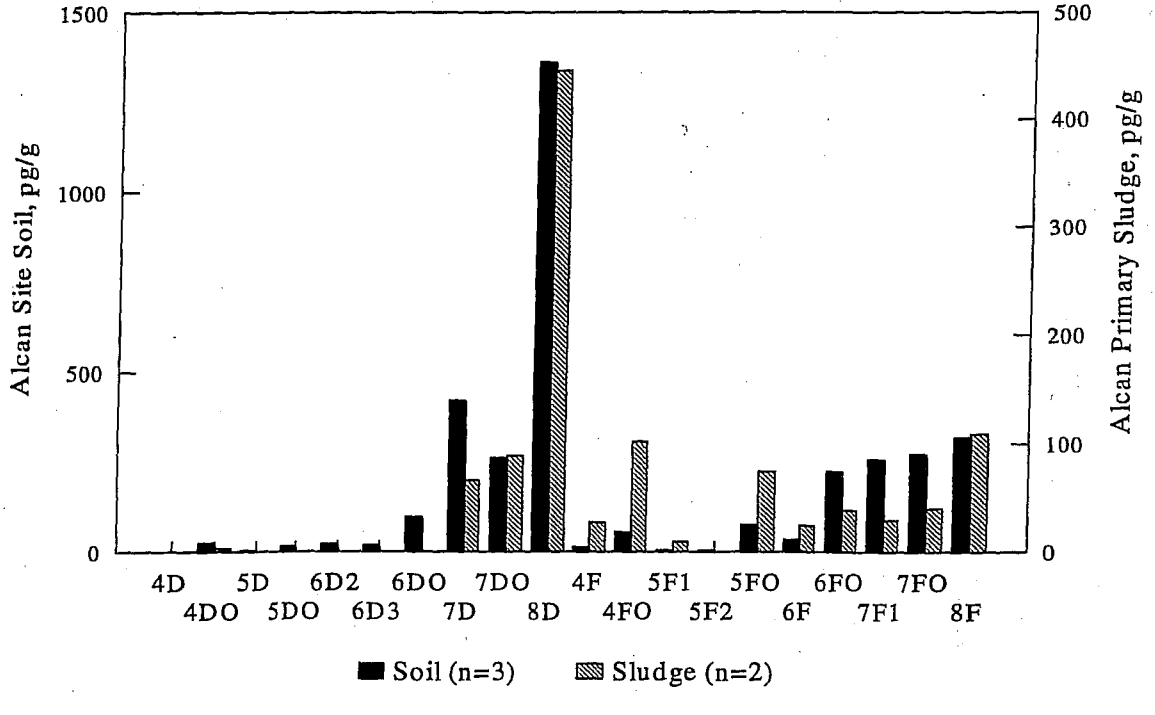


Figure 13-2. Congener profiles for soil and sludge samples from the Alcan smelter.

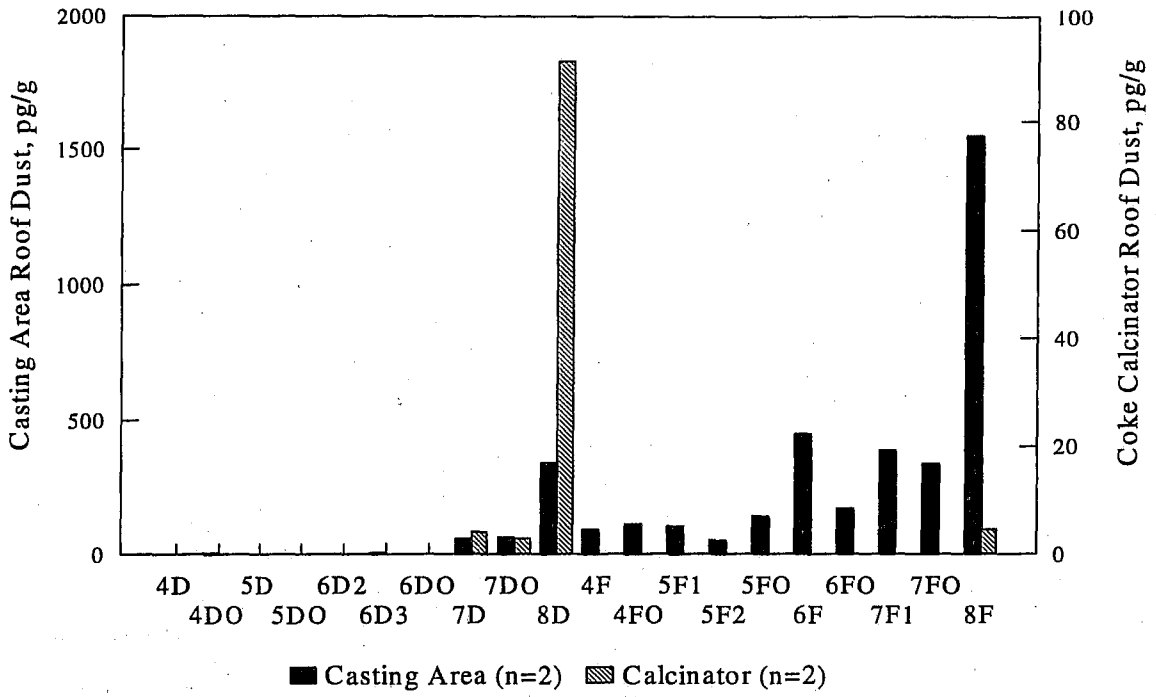


Figure 13-3. Congener profiles for roof dust from the casting area of the Alcan smelter.

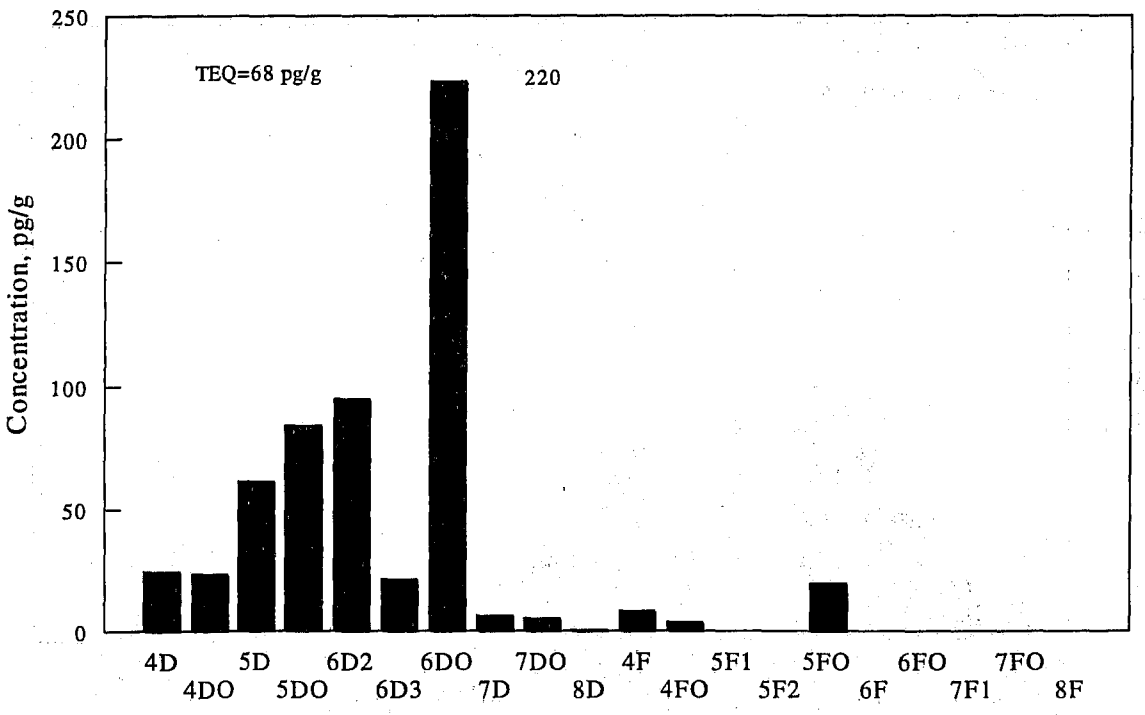


Figure 13-4. Kitimat congener profiles for 1989 (n=8).

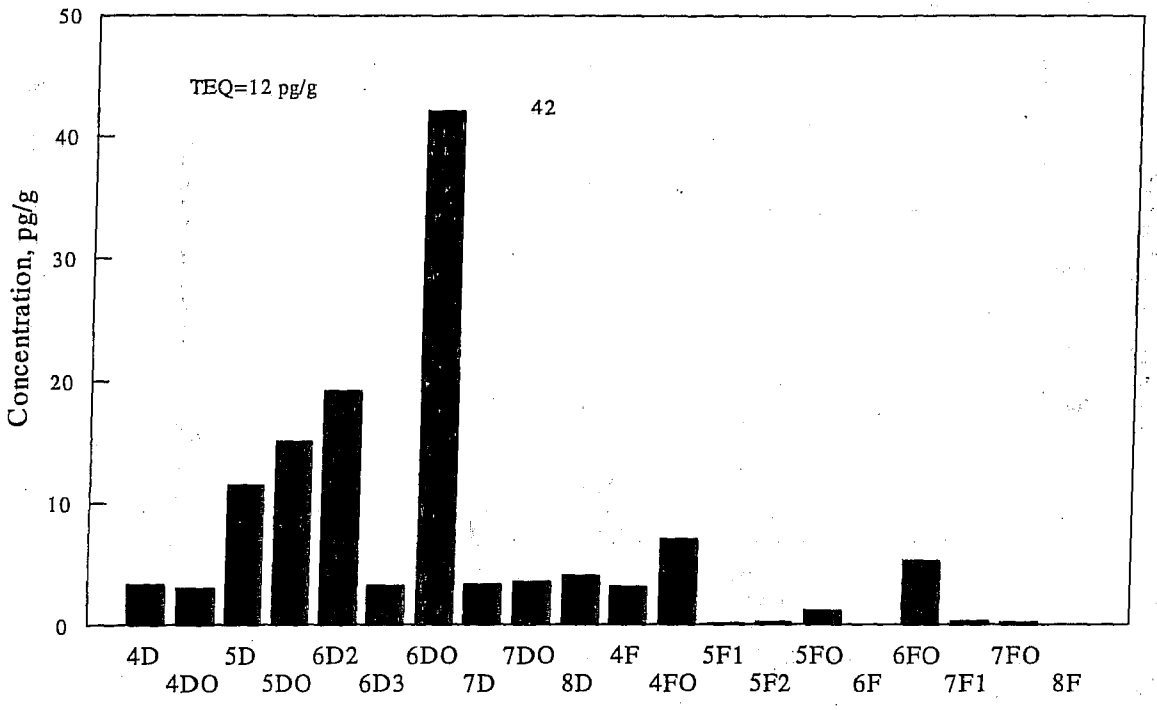


Figure 13-5. Kitimat congener profiles for 1991 (n=7).

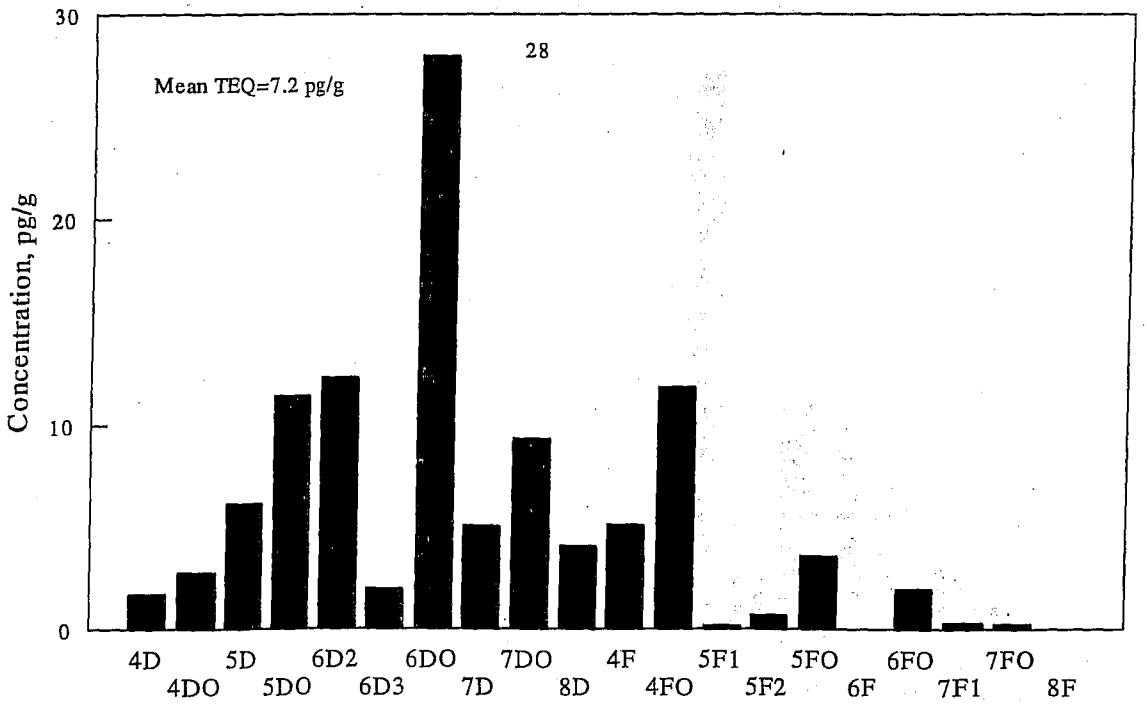


Figure 13-6. Kitimat congener profiles for 1992 (n=41).

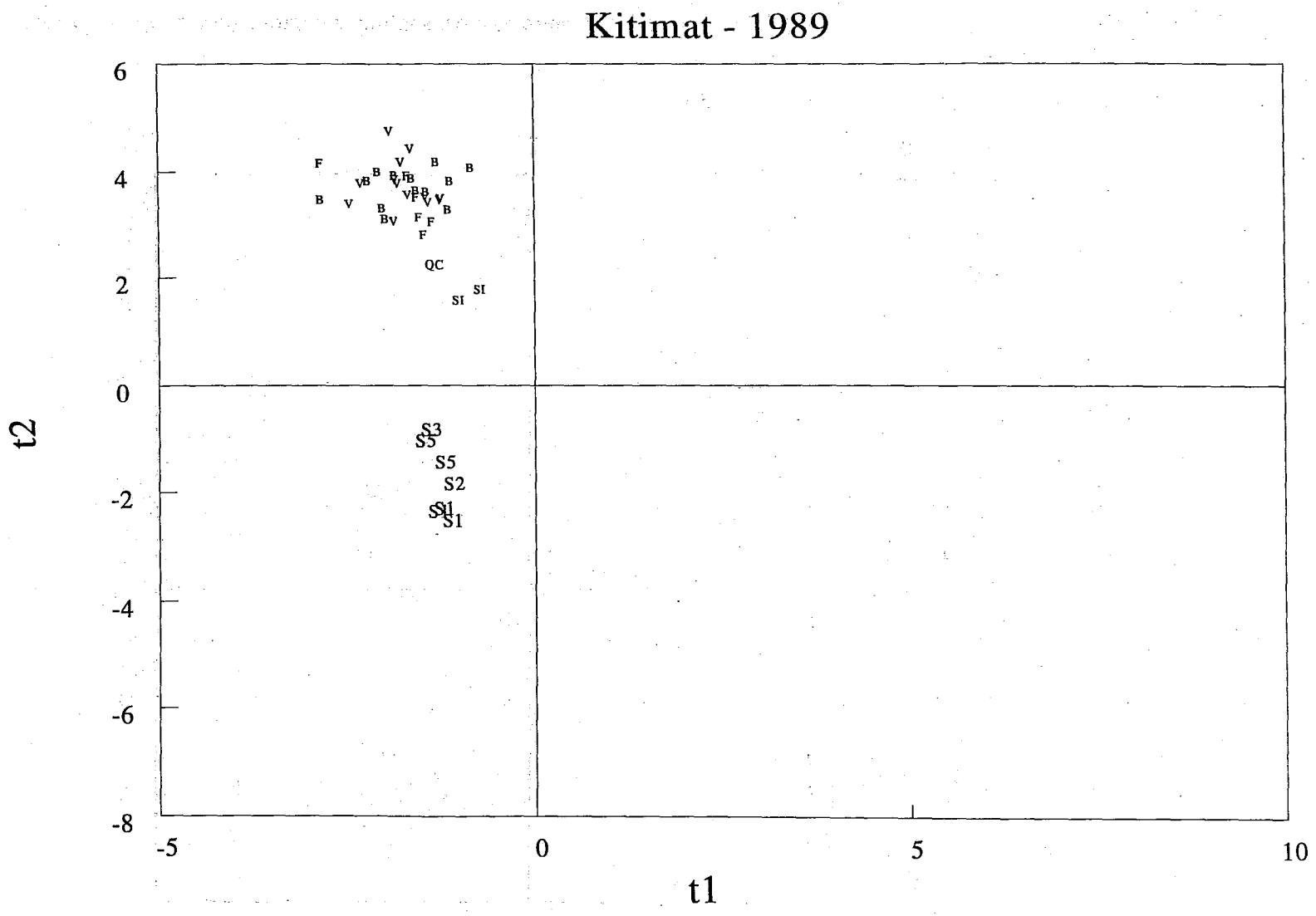


Figure 13-7. PCA projections for samples from Kitimat in 1989.

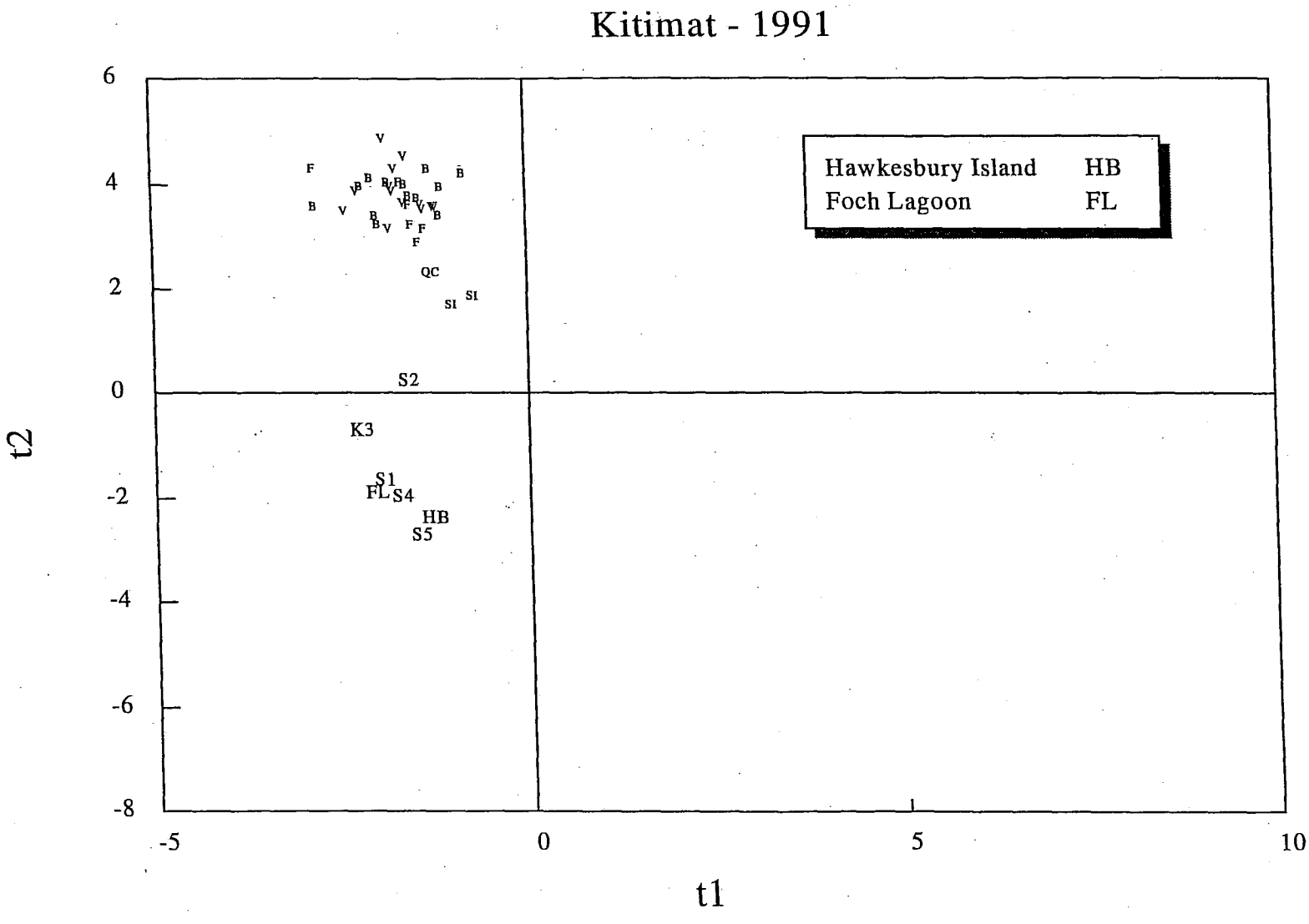


Figure 13-8. PCA projections for samples from Kitimat in 1991.

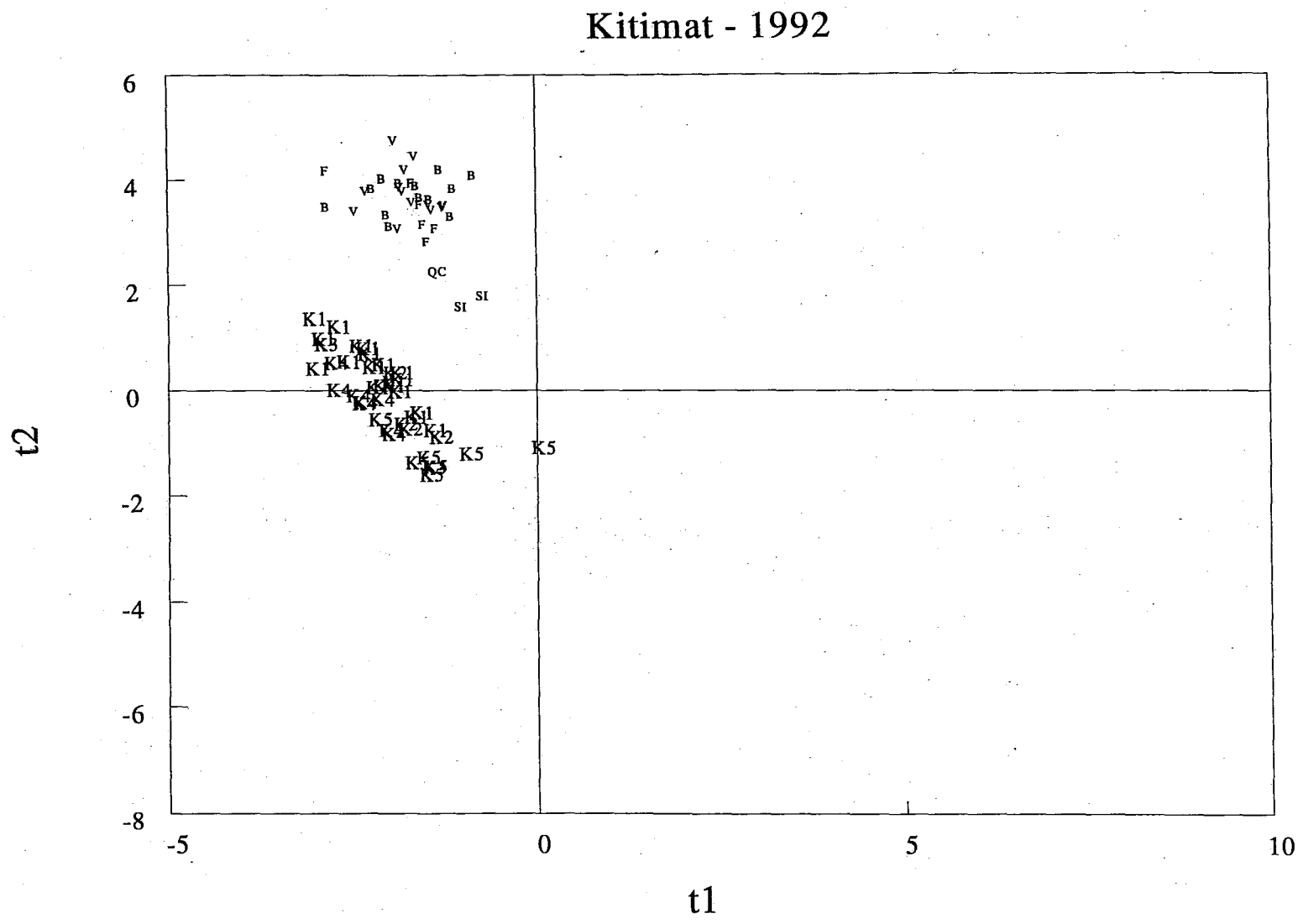


Figure 13-9. PCA projections for samples from Kitimat in 1992.

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14. CONCLUSIONS

PCA is a valuable method for exploring trends and defining relationships in the large sample set of chlorinated dibenzo-*p*-dioxin and dibenzofuran data for samples of crab hepatopancreas that have been collected from B.C. coastal sites. PCA has proven to be particularly useful for classifying samples according to principal source.

- Variations in the proportions of TCDFs from chlorine bleaching and HxCDDs derived from pentachlorophenol have produced striking differences between the PCDD/F profiles of crabs from different mill sites.

- Changes in mill process chemistry since 1987 have produced dramatic improvements. In most cases both the proportion of the toxic 2,3,7,8-chlorinated congeners and the overall chlorinated dibenzo-*p*-dioxin and dibenzofuran concentrations have decreased markedly in crabs collected from the mill sites.

- Mill related TCDFs have been removed faster than the HxCDDs: one effect is that crab composition profiles and PCA projections have become more similar over time at the mill sites.

- Crab samples from B.C.'s harbours have lower proportions of the toxic 2,3,7,8-chlorinated congeners, but have shown less change over time.

- PCA results suggest that differences in feeding behavior may have reduced the toxic load for crab species other than Dungeness crab. It is suggested that whenever possible a single species — Dungeness crab — should be used for the mill monitoring programs.

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