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PAHs in Saint John Harbour sediments - an evaluation of 1996-1999 results

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ABSTRACT

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There are good linear relationships between the sum of concentrations of fifteen PAHs (SPAH) and fluoranthene (Flu): SPAH (mg/kg) = 0.17 + 4.62 * Flu(mg/kg), and between fluoranthene and pyrene, phenanthrene, and benzo(*a*)anthracene. These relationships together with Principal Component Analysis (PCA) of PAH profiles (concentrations expressed in percent) are used to judge the reliability of the data from eight sites in the Saint John Harbour and Courtenay Bay. The expected interlaboratory relative standard deviation (RSD) of SPAH in the 1 mg/kg range is about 100%. Of the 180 samples analyzed from 1996-99, four exceed the guideline of 2.5 mg/kg and an additional four are on the borderline. It is suggested that a single number is not a realistic guideline. Better quality control of the analyses, as well as a better sampling protocol, are needed.

RÉSUMÉ

Zitko, V. 1999. PAHs in Saint John harbour sediments - an evaluation of 1996-1999 results. Can. Tech. Rep. Fish. Aquat. Sci. 2290: iii + 48 p.

Il existe de bonnes relations linéaires entre la somme des concentrations de quinze HAP (SPAH) et de fluoranthène (Flu) [HAP (mg/kg) = 0,17 + 4,62 * Flu (mg/kg)] et entre les concentrations de fluoranthène et de pyrène, de phénanthrène et de benzo(a)anthracène. Ces relations, couplées à une analyse des composantes principales (ACP) des profils HAP (concentrations exprimées en pourcentage), sont utilisées pour déterminer la fiabilité des données recueillies à huit endroits dans le port de Saint John et dans la baie Courtenay. Le coefficient de variation interlaboratoire probable des HAP s'approchant de 1 mg/kg se situe à environ 100 %. Des 180 échantillons analysés de 1996 à 1999, quatre montraient des concentrations au-dessus des cibles de 2,5 mg/kg, tandis que quatre autres se situaient à la teneur autorisée. Un chiffre unique ne semble pas être une limite réaliste. Un contrôle plus étroit de la qualité des analyses, ainsi qu'un meilleur protocole d'échantillonnage, seraient plutôt indiqués.

INTRODUCTION

Aquatic sediments are a major sink for polynuclear aromatic hydrocarbons (PAHs), as well as a source of these compounds for bottom-dwelling biota. As a consequence, the concentration of PAHs in sediments attracts considerable attention (see, for example, Domine et al. 1994, Maedor et al. 1995, Naes et al. 1995, Simpson et al. 1996, Zepp and Macko 1997, Zitko 1993). In addition, PAHs may be released from sediments by dredging and dredge spoil dumping and the environmental impact of these operations is regulated by national and international guidelines. The current Canadian guideline for total PAHs (SPAH) is 2.5 mg/kg of dry sediment.

A large number of sediment samples from the Saint John harbour and vicinity have been analyzed for PAHs, primarily as a requirement for environmental impact assessment of maintenance dredging and dumping. Data from 1990-93 have been summarized by Lindsay and Zitko (1996). The data set consisted of 317 samples. The same set of PAHs was not measured in all samples, but all samples had the measurement of thirteen PAHs in common. Except for the five most common PAHs (phenanthrene, fluoranthene, pyrene, benz(*a*)anthracene, and chrysene), less than half of the samples contained PAH levels measurable by the reporting laboratories. In over 80% of the samples, SPAH for the thirteen reported PAHs was less than 2.0 mg/kg. The relative composition of the PAH mixtures corresponded to a general industrial fallout, with some contribution of petroleum sources in certain areas. Absent were fluoranthene-rich PAH mixtures, reported for sediments from parts of the Gulf of Maine (Zitko 1993, Zitko 1996).

The objective of this work was to evaluate the data obtained since 1993, to determine correlations between PAH concentrations and sediment characteristics (total organic carbon (TOC) and grain size), and to assess potential errors in the data, particularly in samples with elevated SPAH values.

PAH DATA

PAH concentrations in sediment samples taken in 1996, 1998, and 1999 for the Maintenance Dredging projects, were provided by Mr. Ed Vye, P.Eng., Saint John Port Corporation. The general sampling sites are indicated in Fig. 1. Only samples containing more than 0.5 mg/kg of total PAHs (sum of 15 hydrocarbons: naphthalene (N), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Fen), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo(*a*)anthracene (BaA), chrysene (Chr), benzofluoranthenes (BFl), benzo(*a*)pyrene (BaP), indeno(*1,2,3cd*)pyrene (ind), dibenzo(*ah*) anthracene (DahA), and benzo(*ghi*)perylene (ghi)) were considered . The 1996 and 1998 results specified benzo(*b+k*) fluoranthene, and also contained benzo(*e*)pyrene. To make the 1999 data compatible, benzo(*e*)pyrene was deleted from the earlier data sets. It is assumed that 'benzofluoranthenes' in the 1999 report mean benzo(*b+k*) fluoranthene.

The 0.5 mg/kg 'cutoff' was chosen to limit the 'noise' inherent in measurements near the limit of detection (0.01 mg/kg dry weight). If some PAHs were reported as non-detectable, half of the detection limit (0.005 mg/kg) was used.

The sample locations, Total PAH (SPAH) concentrations, and sampling years are listed in Appendix 1 consecutively, and in Appendix 2 according to sites (the approximate sampling sites are shown in Fig. 1). Numbers following 's' in the second column of Appendices 1 and 2 are the original sample numbers in the reports submitted to the Saint John Port Corporation. Appendix 1 also contains the expected relative standard deviations (RSDs) of the SPAH values, calculated according to Horwitz and Albert (1997) and taking into account the propagation of errors during the summation. RSDs are estimates of the expected interlaboratory standard deviations. This aspect of the data is discussed later in the Analytical Uncertainty section. Appendices 3 and 4 list the concentrations of the individual PAHs, in mg/kg dry sediment, and in %, respectively.

GENERAL OBSERVATIONS

There are twelve samples with SPAH>2.0 mg/kg among the eighty-two samples listed in Appendices 1- 4. Only sites: Berth #1, #3, #11, and #12, the Potash Terminal, and the one sample from the Long Wharf Slip do not include samples with SPAH>2.0 mg/kg.

The abundance of PAH in the samples generally decreases in the order fluoranthene > pyrene > phenanthrene > benzofluoranthenes > chrysene > benzo(a)anthracene... (Appendices 3, 4). The first three hydrocarbons account for at least 40% of the total PAH and the first six amount to about 70% of the total. Means and other statistical characteristics of the data are presented in Appendices 5 and 6 and a brief summary is in Table 1.

Table 1. A brief overview of PAH concentrations and their variability (cv= coefficient of variation, 100*std/mean, SPAH=sum of PAH concentrations).

PAH	Mean mg/kg	std	cv	Median mg/kg	Max mg/kg	Min mg/kg	Scaled mean,%	Scaled median, %
N	0.03	0.05	149	0.02	0.43	0.01	2.61	2.63
Acy	0.01	0.01	102	0.01	0.06	0.01	0.56	0.66
Ace	0.02	0.04	178	0.01	0.36	0.01	1.87	1.32
Fen	0.03	0.04	113	0.02	0.25	0.01	2.50	2.63
Phe	0.18	0.20	114	0.11	1.50	0.05	13.63	14.47
Ant	0.06	0.07	118	0.03	0.39	0.02	4.68	3.95
Flu	0.25	0.28	116	0.14	1.83	0.06	18.89	18.42
Pyr	0.21	0.28	137	0.11	2.11	0.05	15.85	14.47
BaA	0.09	0.10	115	0.05	0.67	0.03	6.92	6.58
Chr	0.10	0.11	111	0.06	0.78	0.04	7.79	7.89
BFl	0.14	0.15	108	0.09	1.11	0.05	10.94	11.84
BaP	0.07	0.08	111	0.04	0.55	0.02	5.50	5.26
ind	0.05	0.05	90	0.04	0.29	0.01	4.12	5.26
DahA	0.01	0.01	130	0.01	0.10	0.01	0.88	0.66
ghi	0.04	0.04	84	0.03	0.20	0.02	3.26	3.95
SPAH	1.30			0.76	10.63	0.38	100	100

In the whole data set, standard deviations (std) are close to the respective means and the coefficients of variation (cv) exceed 100% and indicate an asymetric, censored on the left distribution. Left censoring is caused by measurements close to detection limits.

The concentrations of fluoranthene and pyrene are highly correlated (Appendix 7). The concentration of fluoranthene is less well correlated with those of chrysene, benzo(*a*)anthracene, and benzofluoranthenes. There is a good linear relationship between the concentration of fluoranthene and SPAH (Fig. 2), and between fluoranthene and pyrene (Fig. 3). A good linear relationship is retained even after removal of fluoranthene concentrations from SPAH. As indicated already by the correlation coefficients, the relationships between phenanthrene or benzo(*a*)anthracene, and fluoranthene are not as good (Fig. 4, 5). Phenanthrene may originate from petroleum-related sources, containing less fluoranthene. Larger analytical errors may be responsible for the poor relationship between benzo(*a*)anthracene and fluoranthene. Nevertheless, all these relationships can be used to detect gross errors or inconsistencies in the data.

It can be easily seen in Fig. 2 that of the eight samples with SPAH > 2.5 mg/kg, five are above the regression line, which indicates that their SPAH values may have been overestimated. By the same reasoning, SPAH values may be underestimated in the three samples with the highest SPAH values (samples 57,12 and 50). Large discrepancies in the concentration of phenanthrene in samples 50 and 57 are immediately obvious from Fig. 4. The concentrations of PAHs are not correlated with TOC and the particle sizes of the sediments (Appendix 7). The average ratios of pyrene to fluoranthene (PF) and of phenanthrene to anthracene (PA) are 0.81 and 3.57, respectively. For comparison, in Boston harbour sediments these ratios are 1.05 and 4.70, respectively. PF values of 1.67, 0.98, 0.68, and 1.11 are reported for gasoline exhaust, street dust, creosote, and #2 fuel oil, respectively. The PA values of the last two sources are 4.13 and 50 (McGroddy and Farrington 1995). These data indicate that PAHs in the Saint John Harbour originate from general industrial and municipal activities. Source allocation based PAH profiles given by Page et al (1999) give the following source distribution: petroleum tar (residue after evaporation of volatile hydrocarbons) 40%, wood burning 36%, human habitation 22%, and creosote 2%.

PAH PROFILES

In the environment PAHs always occur in mixtures whose composition may identify the source, such as general combustion, creosote, petroleum products, street runoff, sewer discharge, etc. The relative proportions of the individual PAHs in the mixtures are usually expressed in percents and are referred to as PAH 'profiles' (Appendices 4-6). The number of PAHs in these profiles ranges from about five to over 20 (15 in the Saint John data). The profiles are usually reported in tables or bar graphs and are difficult to review (see, for example, Appendix 4). The comprehension of the profiles is facilitated by mathematical techniques such as the Principal Component Analysis (PCA, Zitko 1994, 1996). PCA selects new coordinates, ***principal components***, formed as linear combinations of the original ones in a way that captures most of the original information. The result are ***score plots*** in which similar samples are located close together and ***loading plots***, which indicate the effects of the original variables on the principal components. PCA does not provide new or additional information. It helps to draw attention to the relationships in the data, which could be found anyway, by a close and time-consuming examination. In other words, PCA projects the multidimensional (5-20 dimensions) data on a space of fewer dimensions (2-3). The projections can be examined visually in ***score plots*** (axes marked 'pc'), in which similar samples are located in close proximity. The effects of the individual PAHs on the score plots can be examined in ***loading plots*** (axes marked 'ev'), in which correlated PAHs are clustered together.

As can be seen from the loading plot of the first two principal components (Fig. 6, ev-1, ev-2), the first principal component separates phenanthrene, acenaphthene, fluorene, fluoranthene, and pyrene from the higher molecular weight PAHs and is not affected by naphthalene and anthracene. The second principal component separates fluoranthene and pyrene from acenaphthene, phenanthrene, fluorene, and anthracene.

Consequently, samples with relatively high concentrations of fluoranthene and pyrene will be located in the left upper quadrant of the score plots pc-2 *vs* pc-1, samples with higher concentrations of acenaphthene, phenanthrene, fluorene, or anthracene will be in the left lower quadrant, and samples with relatively elevated levels of the higher molecular weight PAHs will be shifted to the right-hand side of the score plots. The third principal component separates particularly naphthalene and acenaphthylene from anthracene (Fig. 7).

The next step in the evaluation of the data is to examine the score plots and to identify samples with unusual PAHs profiles. To prevent overcrowding of the plots, only samples from given areas are identified by numbers (refer to the first column in Appendices 1 and 2). The positions of other samples are marked by dots, to keep in perspective the relation of given samples to the whole data set.

RODNEY SLIP AND BERTH #3

The three Rodney Slip samples (Fig. 8, samples 2, 20, and 56) are from 1996, 1998, and 1999, respectively and are from approximately the same area at the east end of Rodney Slip. The Berth #3 samples are also from the east end of the berth, slightly more inside the site. For the whole site, sample '2' is an outlier, with relatively higher concentrations of fluoranthene and pyrene (Fig. 8). The remaining six samples form two clusters in the pc-1 & pc-2 plane, but do not cluster vertically (in the pc-1 & pc-3 plane, Fig. 9). The PAH profile of the sample with the highest total PAH concentration (sample 56, SPAH=4.75 mg/kg) has a profile usual for this site. The sample 56 area and its close vicinity should be sampled on a finer grid to delineate the extent of contamination.

RODNEY MARGINAL

Except for samples 11 and 58, the SPAH values, predicted from the concentration of fluoranthene, are in good agreement with the observed ones. On the other hand, PAH profiles from the Rodney Marginal site are the most widely scattered profiles encountered in this study. The samples 4 and 5, taken in 1996, and the sample 57, taken in 1999 come from the 'Bay' end of Rodney Marginal. The samples 22 and 60 were taken near the centre, in 1998 and 1999, respectively. The sample 60 is from the vicinity of a sewer outfall. The samples 21 taken in 1998, and samples 58 and 59, taken in 1999 come from about the inland fourth of Rodney Marginal. The remaining three samples, 3, 11, and 57, were collected in 1996, 1998, and 1999, respectively at the inland end of Rodney Marginal. For the whole site, samples 57, 22a, and 59 are outliers (Fig. 10, 11). The sample 57 is unusually rich in phenanthrene (see also Fig. 4) and acenaphthene, the sample 59 in benzo(*a*)pyrene and indeno(1,2,3cd)pyrene. The sample 22 is enriched in naphthalene. On the other hand, sample 22a, from the vicinity of sampling stations 22 and 60, is enriched in the higher-molecular-weight PAHs and so is the sample 59

Since the PAH profiles from this site are so varied, it was of interest to look at the profiles of all samples taken at this site (Fig.12, 13). On this scale, sample 59 and seven samples from previous years are outliers. The very high variability of PAH profiles in samples from this site suggests that a re-sampling of the whole site or at least the vicinity of stations 12 and 57 may have to be carried out, on a finer grid.

Four additional samples from a dredging block adjoining that of sample 57 have been analyzed recently. The results, together with others from the same area, not considered previously because of their SPAH<0.5, are summarized in Appendix 10. It is worth noting that the estimates of SPAH (see Fig. 2) are in good agreement with the measured values, except for sample 58. The dredging sample grid is shown in Appendix 11.The additional samples were taken in block 4, in the area marked approximately by 'X'. As can be seen from Appendices 10 and 11, there is a very high spatial variability of PAH concentrations. The variability in PAH profiles is impossible to judge in this case, because of the large proportion of 'not detectable' concentrations, set arbitrarily at 0.005 mg/kg. The spatial variability of PAH concentrations makes it necessary to carry out sampling on a well-defined grid, under statistical control (see for example Gilbert 1987).

BERTH #1 AND LONG WHARF SLIP

These two sites were combined because of the small number of samples (Berth #1: samples 9 and 53, Long Wharf Slip: sample 15). The levels of PAHs in all three cases are quite low, but both Berth #1 samples appear to be outliers from the whole PAH profile population (Fig. 14, 15). The sample 9 is so high in anthracene that the value is almost certainly in error (unless anthracene as a chemical was shipped and spilled at the site). The sample 53 has the highest reported concentration of benzo(*ghi*)perylene and that also may be an analytical error.

BERTH #11 AND #12

PAH profiles from the Berth #12 site form a fairly tight cluster (Fig. 16, 17). The only sample with SPAH > 0.5 mg/kg from Berth #11 (6) is in comparison to samples from Berth #12 enriched in fluoranthene and pyrene and resembles more samples 4, 5, and 12 from the 'Bay' end of Rodney Marginal (Fig. 10).

LOWER COVE

The PAH profiles at this site have three outliers (samples 24, 65 and 18, Fig. 18, 19). The sample 24 is relatively enriched in acenaphthene, which is unusual, sample 65 contains relatively elevated levels of phenanthrene. Sample 18 contains an unusually high concentration of naphthalene. This may be an analytical error or an indication of a recent contamination by creosote.

COURTENAY BAY

The PAH profiles of samples from this site indicate three outliers (samples 29, 25, and 75, Fig. 20, 21). The sample 29 is relatively enriched in phenanthrene, fluoranthene and pyrene, the sample 75 in naphthalene, and the sample 25 in dibenzo(*ah*)anthracene, which probably is an analytical error. The PAH profile of the sample 30, which has the highest SPAH value (3.25 mg/kg), is within the cluster of profiles of this site. This sample was taken in 1998. Re-sampling of the same area (CB4) in 1999 yielded sample 75, with a very different PAH profile, as mentioned above. The sample 29 was collected in 1998 in an adjacent area and has also an unusual PAH profile. The CB4 area should re re-sampled on a finer grid.

POTASH TERMINAL

The PAH profiles of this site show one outlier (sample 72, Fig. 22, 23), with highly unusual relative concentration of anthracene. An analytical error is suspected. Otherwise, the PAH profiles of this site form a tight cluster and the samples cover reasonably well the length of the terminal.

CANAPORT

The sample 50 may be an outlier (Fig. 24, 25) and is the only sample in the whole set that contains relatively more pyrene than fluoranthene, which is unusual (see also Fig.3, 4). The samples 47 and 48 have identical profiles and a transcription error is suspected.

PROFILES OF SAMPLES WITH SPAH>2.0 MG/KG

With the exception of samples 57, 5, and 58, the remaining samples form a cluster in the pc-1 & pc-2 plane, but the pc-1 & pc-3 plane projection hints at possible additional outliers (samples 12, 18, and 56, Fig. 26, 27). The samples 57 and 58 are unusually high in phenanthrene. In addition, sample 58 also contains a very elevated relative concentration of anthracene. The sample 18 has an elevated concentration of naphthalene. Fluranthene and pyrene are responsible for the 'outlier' status of sample 12.

UNCERTAINTY IN THE MEASUREMENT OF PAHS IN SEDIMENTS

As can be seen from Appendix 8, the SPAH values for standard reference sediments have a confidence limit of about 30 % (from 23-45 mg/kg for HS-5 and from 18-38 mg/kg for EC-2), and one can assume that the probable error would be about 15%. This is the degree of uncertainty found at PAH concentrations at least one order of magnitude higher than those encountered in the sediments in the Saint John Harbour area. The inter-laboratory relative standard deviation (RSD) of any chemical measurement depends on the concentration of the measured substance and increases from 2% at 100% concentration to 45 % at 1 μg/kg (ppb) concentration. The SPAH calculation includes the summation of 15 measurements, each contributing to an overall RSD. The RDSs of the standard reference sediments reach 60-70% when one takes into account error propagation during the addition of individual PAH concentrations to yield SPAH. Lower PAH concentrations in the Saint John Harbour, relative to those in the stardard reference sediments cause a higher uncertainty, with Horwitz and Albert RSDs of the order of 100% (Appendix 1). Consequently, the probable error of the PAH concentrations in the harbour may be about 25%. On this basis, if the current guideline is 2.5 mg/kg, the 'probable error' interval, in which a deviation from the guideline cannot be determined by a single measurement, may extend from 2-3 mg/kg..

This is a considerable uncertainty, which must be taken into account when evaluating concentrations of PAHs in sediments. Instead of a single number guideline, such as for example 2.5 mg/kg, one should consider an interval, for example from 2-3 mg/kg to be within the guideline. This should be done in conjunction with an assessment of the quality of the data, based on the examinations of relationships among individual PAHs (see for example Fig. 2-5), and of the whole PAH profiles. Thus for instance, samples 11, 19, and 30 (Fig. 2) could be considered 'in compliance', a repeated analysis could decide the status of sample 18, and samples 56, 57, 12, and 50 clearly exceed the guideline and additional sampling and analyses of their respective areas is warranted. Corroborating information could also be obtained by reporting the concentrations of additional hydrocarbons, such as C1-, and C2-naphthalenes and phenanthrenes, paraffins, cycloparaffins, etc. If the analyses are carried out by gas chromatography-mass spectrometry (GCMS), these should be readily available. More sampling and analyses would be required for samples exceeding the 'tolerance' interval, since sampling causes an additional uncertainty. Sediments are generally very heterogeneous and it cannot be expected that one relatively small sample can adequately describe sediment in a 600-m^2 'dredge cell'. In an ideal world, sampling should be carried out according to a statistical design (see for example Gilbert 1987, Keith 1988), with known probabilities of finding 'hot spots'. A 10m-square grid may be the ideal solution. As a compromise, it seems that at least two samples should be taken in a 'dredge cell' and cheaper rapid screening methods should be developed. The data, particularly from Rodney Marginal, indicate that it may be more important to analyze a larger number of samples by a faster screening method. The depth of sampling (vertical profile of PAHs in the sediment) and the depth of dredging should also be taken into consideration. The question that arises in this connection is – is the guideline SPAH concentration in an analytical sample the criterion for the permissibility of dredging, or should the guideline SPAH concentration apply to the average concentration in the dredged material?

QUALITY CONTROL

It appears that only one analysis of a known standard reference sediment per year was done in the years 1996, 1998, and 1999, and there is no evidence of comments or corrective actions when considerable differences occurred between certified and determined concentrations (Appendix 9). Standard reference samples must be analyzed more frequently to ensure that the analyses are under 'statistical control' (Keith 1991). In addition, the samples must include uncompromised reference materials and latent duplicates to obtain an unbiased picture of the performance of the analytical laboratory.

In any case, it should be a standard practice to re-analyze all samples with SPAH >2.0 mg/kg, first, by a duplicate injection. If the results are in good agreement, then proceed with duplicate extraction, and, when these results are in good agreement, then re-sample the area on a finer grid.

CONCLUSIONS

- Of the one hundred and eighty samples analyzed for PAHs in the years 1996, 1998, and 1999, 82 samples contained total PAH concentration (SPAH) exceeding 0.5 mg/kg, and of these, twelve exceeded 2.0 mg/kg (elevated SPAH).

- The largest contiguous area of elevated SPAH was Lower Cove in 1998. SPAH values were lower in 1999.

- PAH profiles and correlations between the main PAHs (fluoranthene, pyrene, benzo(*a*)anthracene and phenanthrene) and between SPAH and fluoranthene are good tools in examining data and may help to indicate patterns, trends, and analytical errors.

- PAH concentrations are not correlated with TOC and particle size distribution in the sediments.

- Samples from Rodney Marginal have the most variable PAH profiles.Since three (56, 57, and 12) of the four samples with highest SPAH values are from this site, additional sampling and analyses should be carried out.

- Sample with the highest SPAH value (sample 50, Canaport, SPAH = 8.77 mg/kg) has a very unusual PAH profile. A survey of the area should be carried out on a finer grid.

- Because of large uncertainties associated with the measurement of PAHs, single-number guideline is not a realistic criterion of compliance.

- The current quality control and assurance procedures are insufficient.

- The sediment sampling protocol is not well developed. Sampling grid should be based on statistical considerations and the relations of guideline concentrations to the volume of dredged material should be defined.

- Areas showing high concentrations of PAH should be routinely re-sampled on a finer grid.

- The development of fast and cheap screening methods, such as the determination of fluoranthene, UV or fluorescence spectrophotometry, should be explored.

- The laboratory reports should be provided both in electronic format and as a hard luation along the lines presented in this report.

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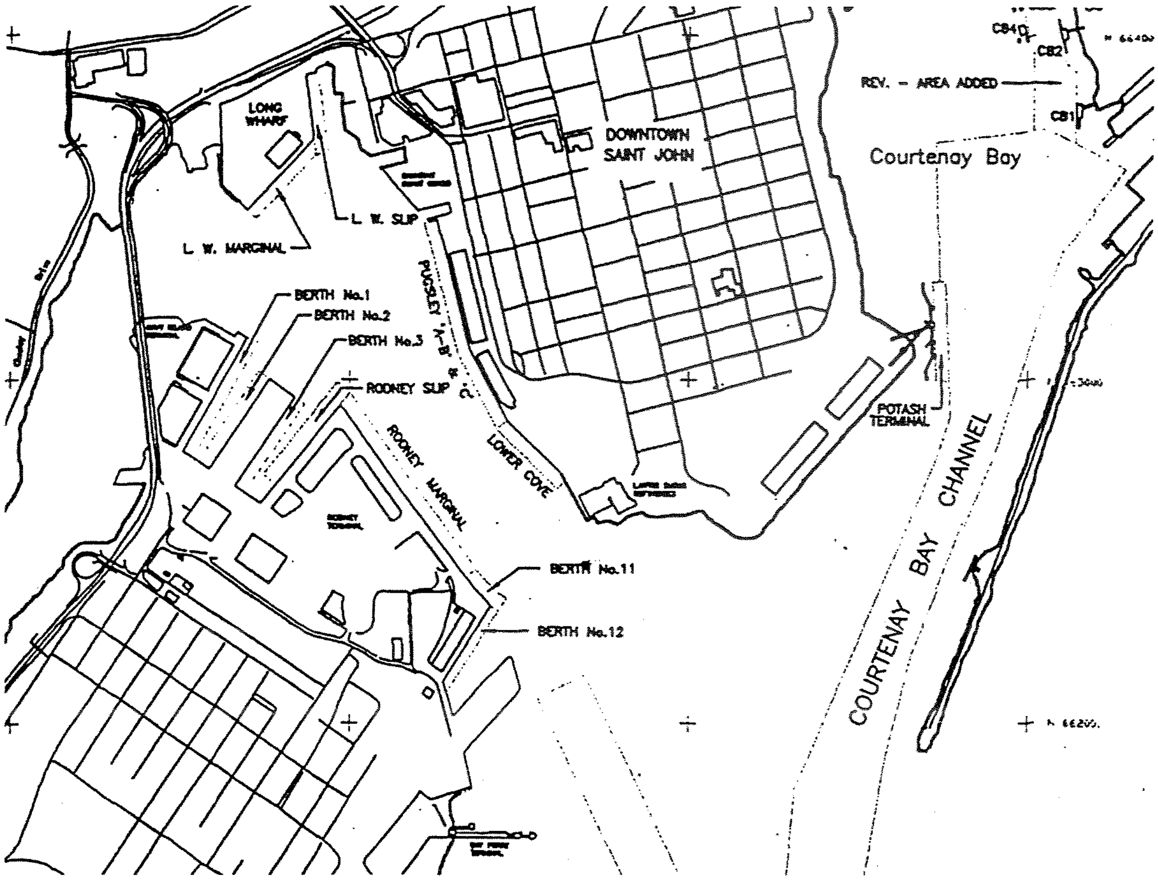

Fig.1 Saint John Harbour and Courtenay Bay with indications of sampling sites

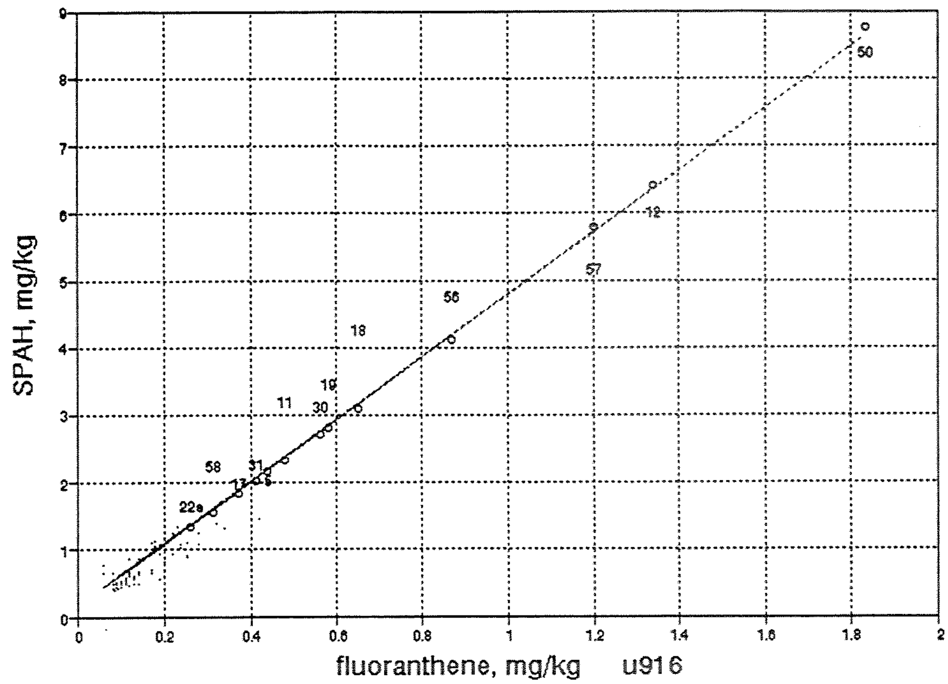

Fig.2 Sum of concentrations of 15 PAHs (SPAH) plotted against the concentration of fluoranthene. Numbers indicate stations with SPAH>1.5 mg/kg, other stations are indicated by dots. Circles show estimated SPAH values when the respective samples are not present in the data set. For information on sampling stations see Appendices 1-4. SPAH [mg/kg] = 0.166 + 4.62 * fluoranthene [mg/kg]. When concentration of fluoranthene are subtracted from SPAH, the relationship becomes SPAH'[mg/kg] = 0.169 + 3.62 * fluoranthene [mg/kg], where SPAH' is (SPAH – fluoranthene).

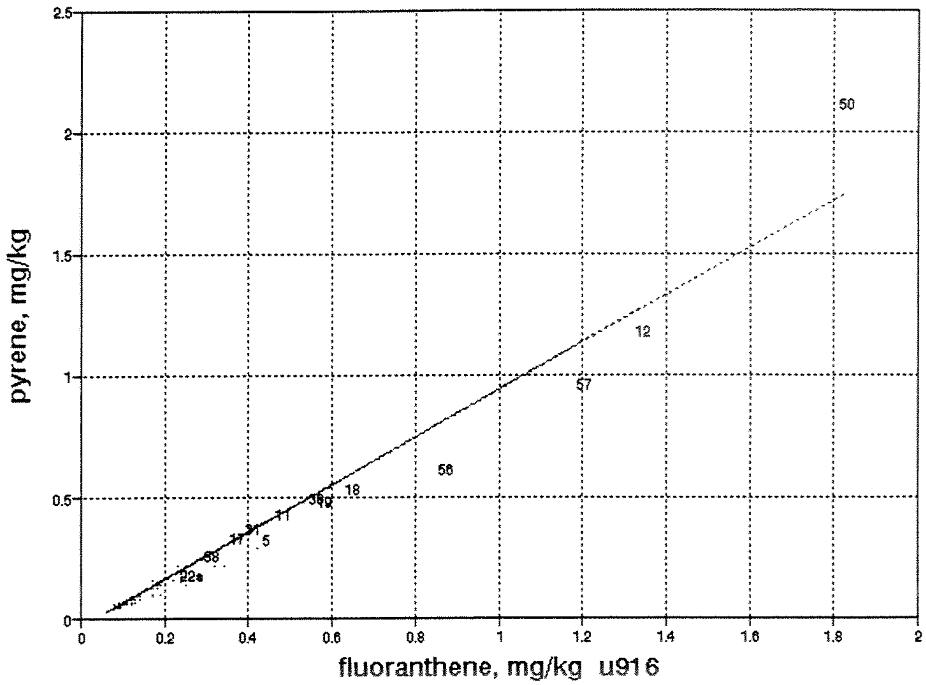

Fig.3 Concentration of pyrene plotted against the concentration of fluoranthene. Numbers indicate stations with SPAH>1.0 mg/kg, other stations are indicated by dots. Circles show estimated SPAH values when the respective samples are not present in the data set. For information on sampling stations see Appendices 1-4.
Pyrene [mg/kg] = -0.03 + 0.97 * fluoranthene [mg/kg]

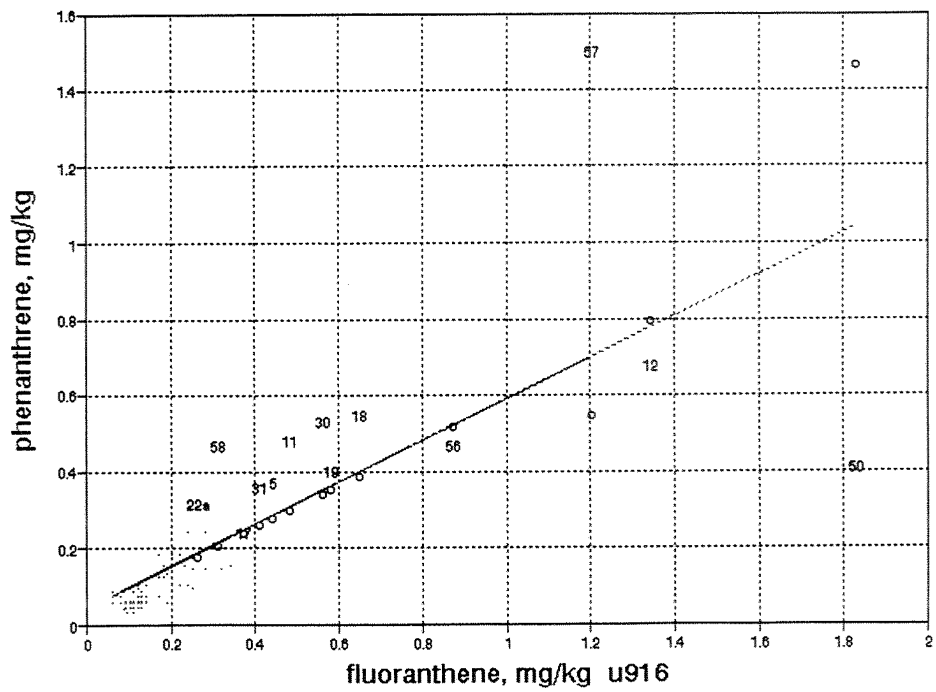

Fig.4 Concentration of phenanthrene plotted against the concentration of fluoranthene. Numbers indicate stations with SPAH>1.0 mg/kg, other stations are indicated by dots. Circles show estimated SPAH values when the respective samples are not present in the data set. For information on sampling stations see Appendices 1-4.
Phenanthrene [mg/kg] = 0.04 + 0.54*fluoranthene[mg/kg]

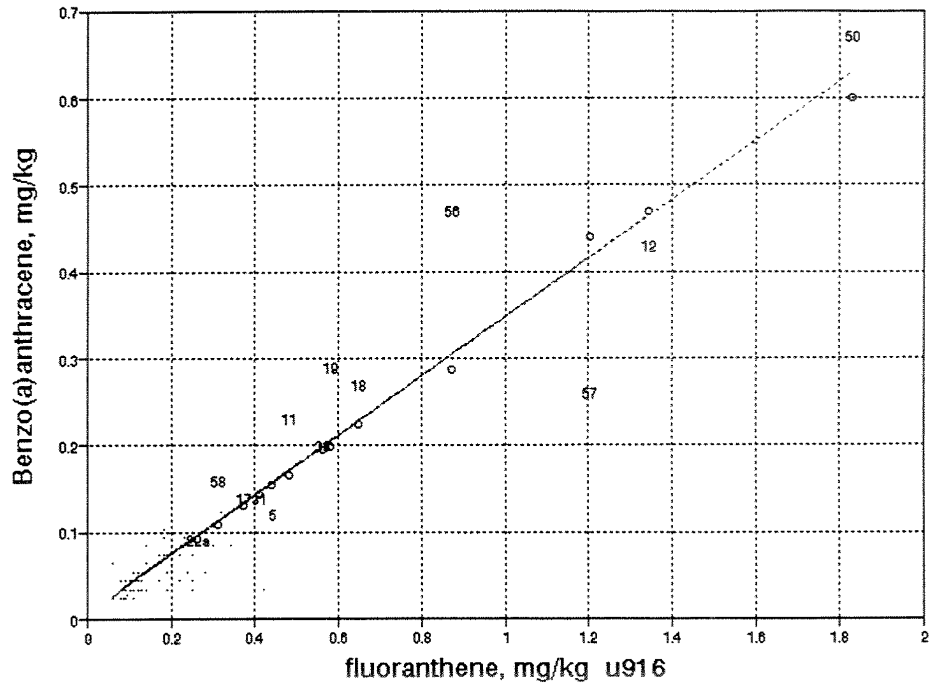

Fig.5 Concentration of benzo(a)anthracene plotted against the concentration of fluoranthene. Numbers indicate stations with SPAH>1.0 mg/kg, other stations are indicated by dots. Circles show estimated SPAH values when the respective samples are not present in the data set. For information on sampling stations see Appendices 1-4.
Benzo(a)anthracene [mg/kg] = 0.01 + 0.34*fluoranthene [mg/kg]

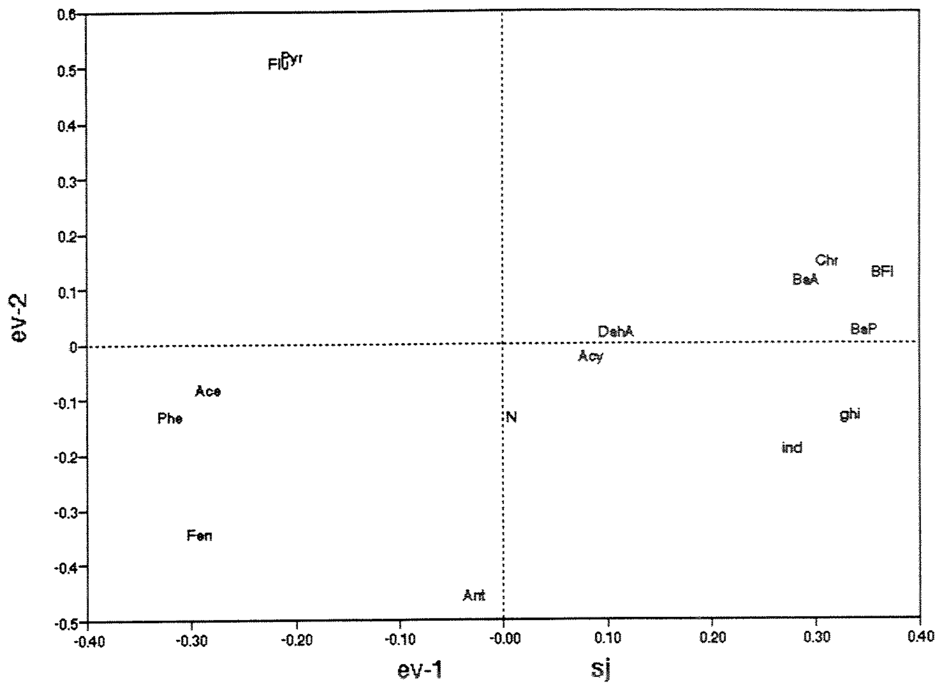

Fig.6 Loading plot showing the effects of the original variables (individual PAHs) on the principal components 1 and 2 (ev-1 and ev-2, respectively). The positions of individual PAHs are indicated by their acronyms: N = naphthalene, Acy = acenaphthylene, Ace = acenaphthene, Fen = fluorene, Phe = phenanthrene, Ant = anthracene, Flu = fluoranthene, Pyr = pyrene, BaA = benzo(*a*)anthracene, Chr = chrysene, Bfl = benzofluoranthenes, BaP = benzo(*a*)pyrene, ind = indeno(*1,2,3cd*)pyrene, DahA = dibenzo(*ah*)anthracene, and ghi = benzo(*ghi*)perylene. Positively correlated PAHs are located close together, negatively correlated PAHs are far apart.

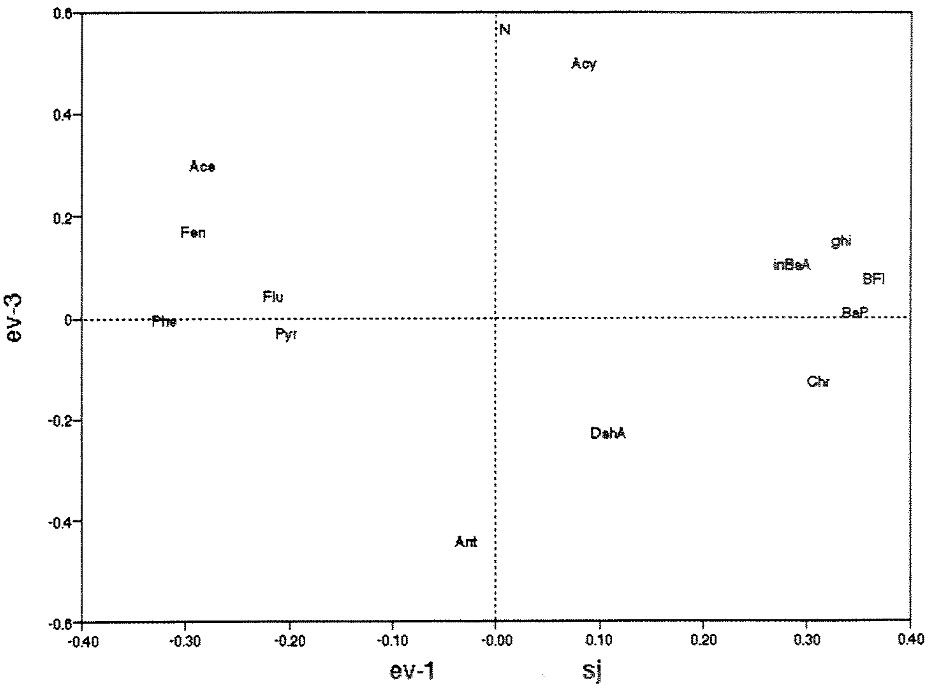

Fig.7 Loading plot showing the effects of the original variables (individual PAHs) on the principal components 1 and 3 (ev-1 and ev-3, respectively). The positions of individual PAHs are indicated by their acronyms: N = naphthalene, Acy = acenaphthylene, Ace = acenaphthene, Fen = fluorene, Phe = phenanthrene, Ant = anthracene, Flu = fluoranthene, Pyr = pyrene, BaA = benzo(*a*)anthracene, Chr = chrysene, Bfl = benzofluoranthenes, BaP = benzo(*a*)pyrene, ind = indeno(*1,2,3cd*)pyrene, DahA = dibenzo(*ah*)anthracene, and ghi = benzo(*ghi*)perylene. Positively correlated PAHs are located close together, negatively correlated PAHs are far apart.

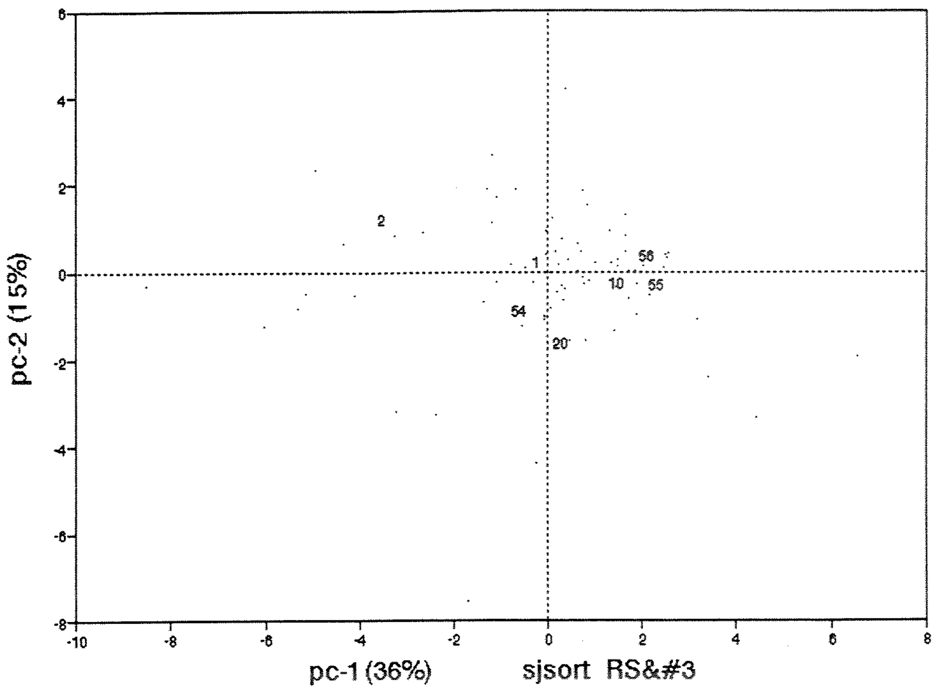

Fig.8 Projection of PAH profiles on the plane of the principal components 1 and 2 (pc-1 and pc-2). Numbers indicate samples from the Rodney Slip and Berth #3 sites, samples from other sites are indicated by dots. Similar samples are located close together. Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

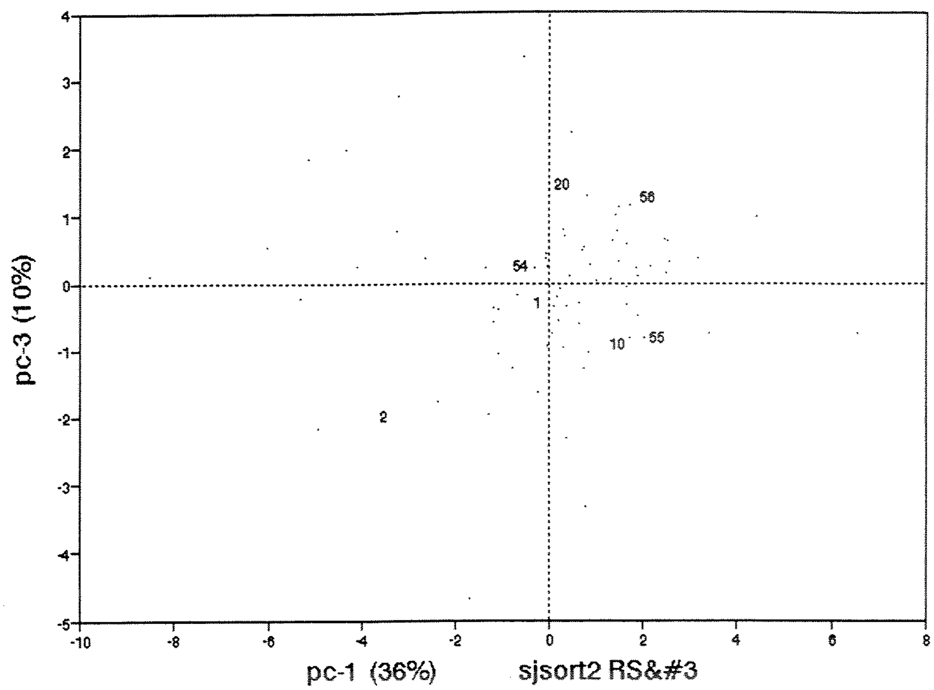

Fig.9 Projection of PAH profiles on the plane of the principal components 1 and 3 (pc-1 and pc-3). Numbers indicate samples from the Rodney Slip and Berth #3 sites, samples from other sites are indicated by dots. Similar samples are located close together. Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

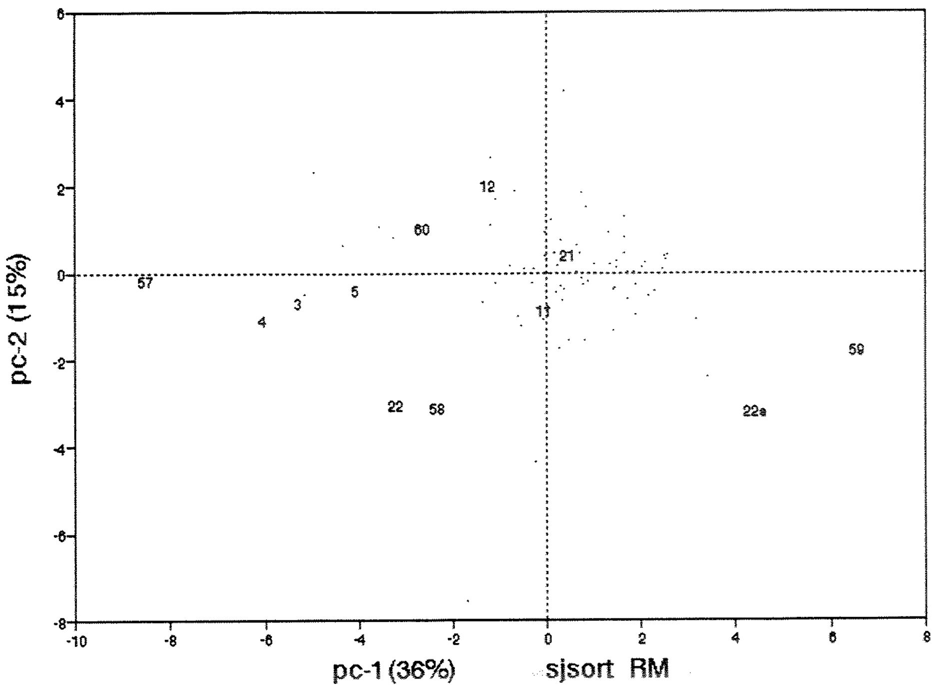

Fig.10 Projection of PAH profiles on the plane of the principal components 1 and 2 (pc-1 and pc-2). Numbers indicate samples from the Rodney Marginal site, samples from other sites are indicated by dots. Similar samples are located close together.
Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4. For information on sampling stations see Appendices 1-4.

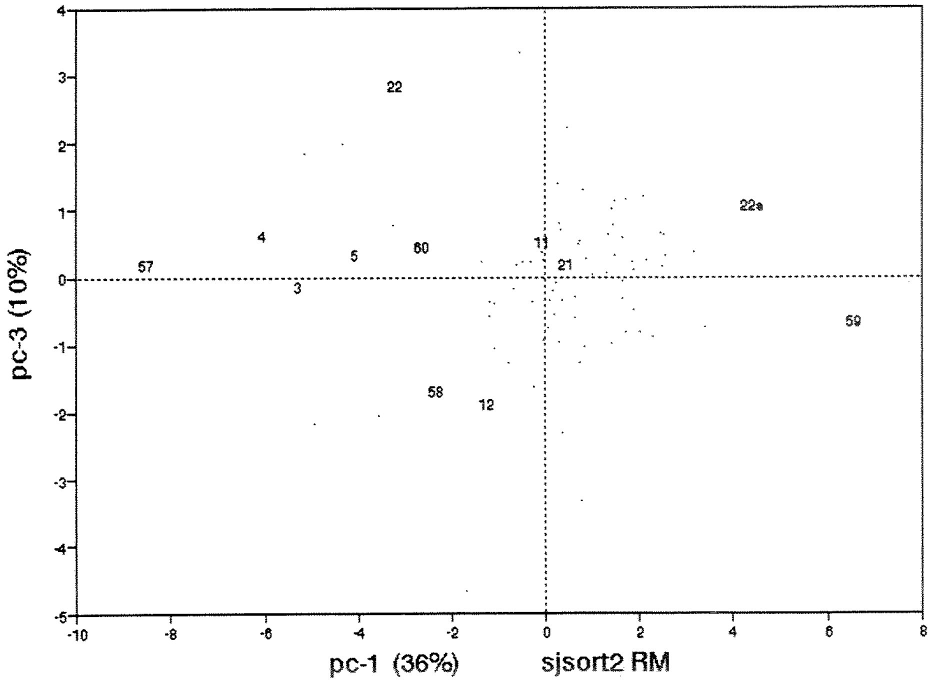

Fig.11 Projection of PAH profiles on the plane of the principal components 1 and 3 (pc-1 and pc-3). Numbers indicate samples from the Rodney Marginal site, samples from other sites are indicated by dots. Similar samples are located close together.
Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

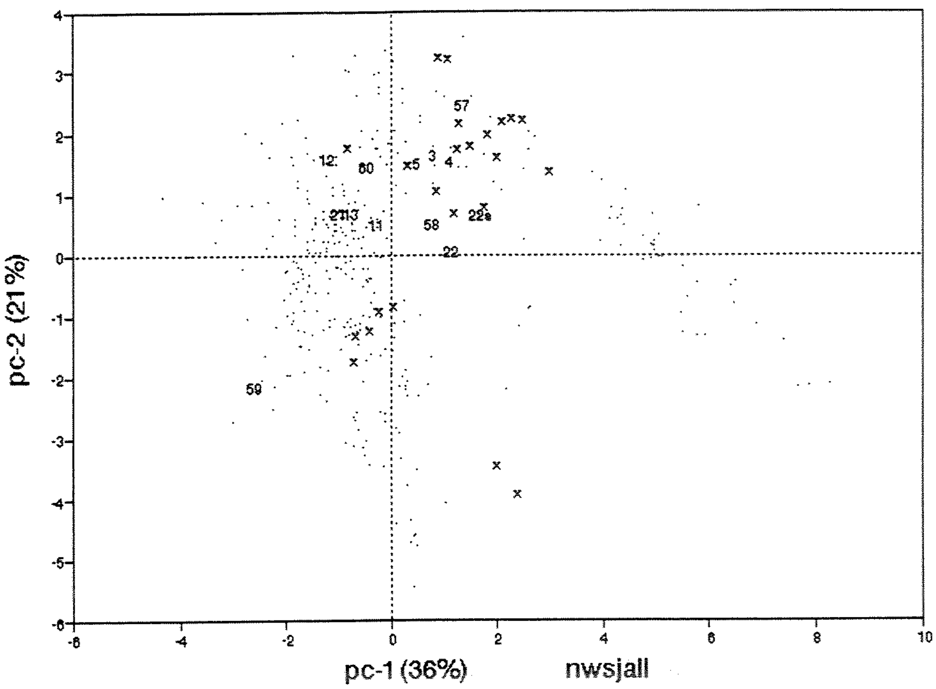

Fig.12 Projection of PAH profiles on the plane of the principal components 1 and 2 (pc-1 and pc-2). Numbers indicate samples from the Rodney Marginal site taken in 1996, 1998, and 1999, and 'x' samples taken in previous years. Samples from other sites collected during this time are indicated by dots. Similar samples are located close together. Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on numbered sampling stations see Appendices 1-4.

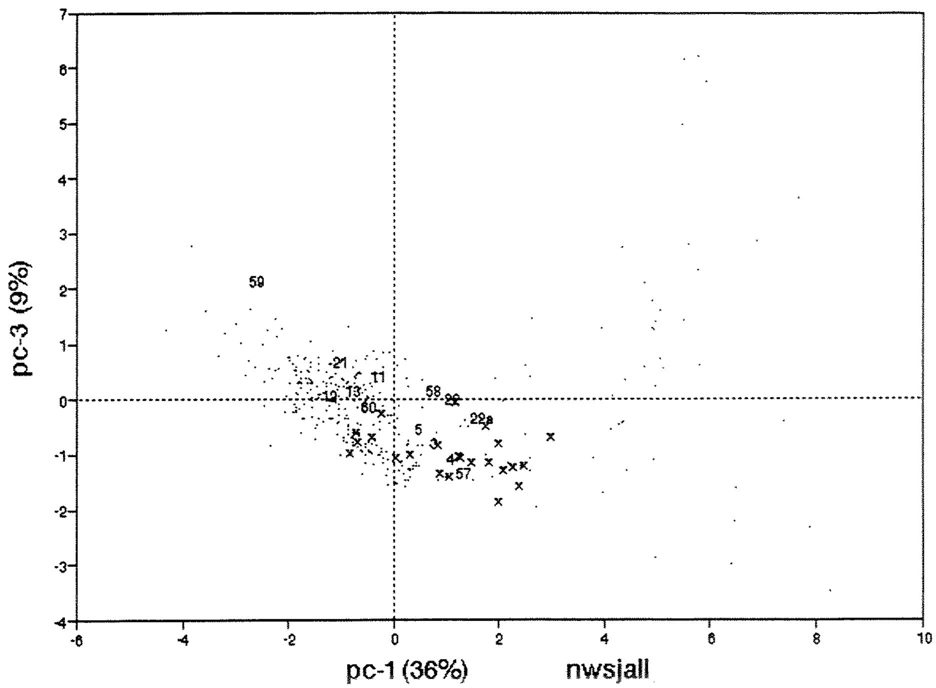

Fig.13 Projection of PAH profiles on the plane of the principal components 1 and 3 (pc-1 and pc-3). Numbers indicate samples from the Rodney Marginal site taken in 1996, 1998, and 1999, and 'x' samples taken in previous years. Samples from other sites collected during this time are indicated by dots. Similar samples are located close together. Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on numbered sampling stations see Appendices 1-4.

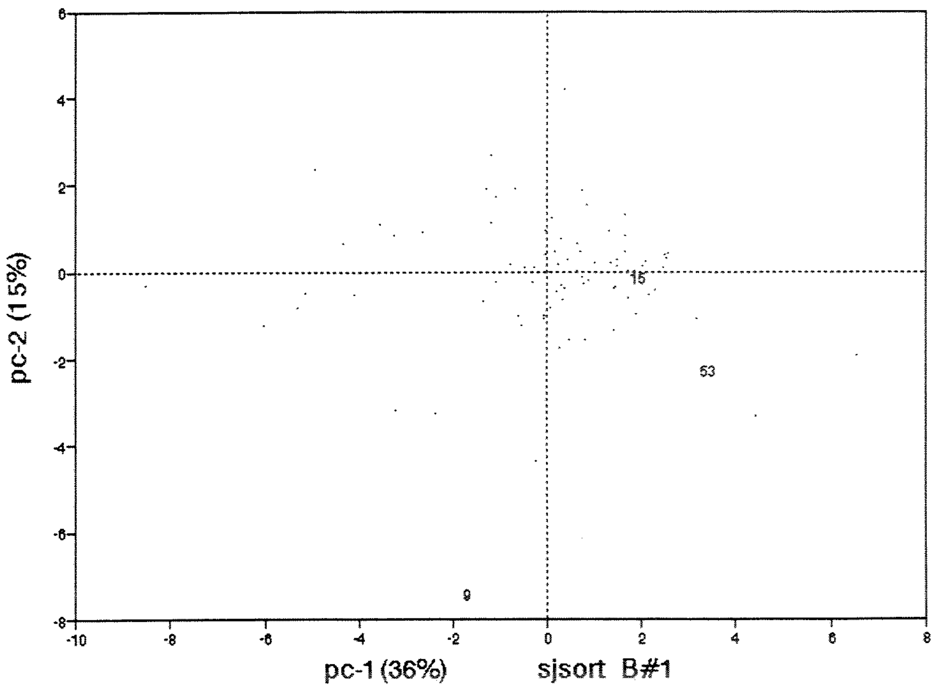

Fig.14 Projection of PAH profiles on the plane of the principal components 1 and 2 (pc-1 and pc-2). Numbers indicate samples from Berth #1 (samples 9 and 53), and from Long Wharf Slip (sample 15), samples from other sites are indicated by dots. Similar samples are located close together.
Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

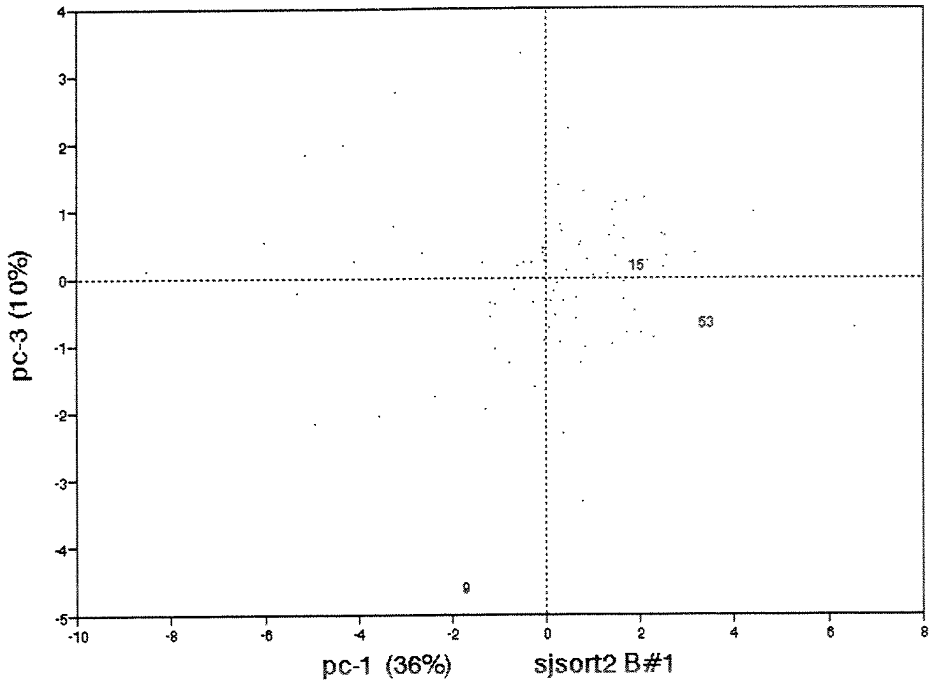

Fig.15 Projection of PAH profiles on the plane of the principal components 1 and 3 (pc-1 and pc-3). Numbers indicate samples from Berth #1 (samples 9 and 53), and from Long Wharf Slip (sample 15), samples from other sites are indicated by dots. Similar samples are located close together.

Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

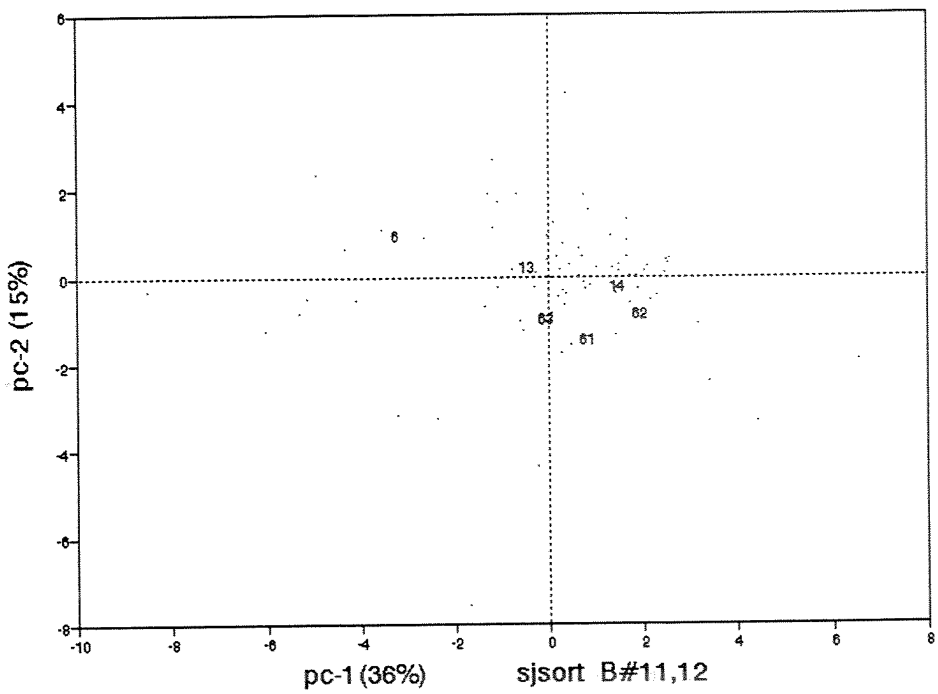

Fig.16 Projection of PAH profiles on the plane of the principal components 1 and 2 (pc-1 and pc-2). Numbers indicate samples from the Berth #11 and Berth #12 sites, samples from other sites are indicated by dots. Similar samples are located close together. Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

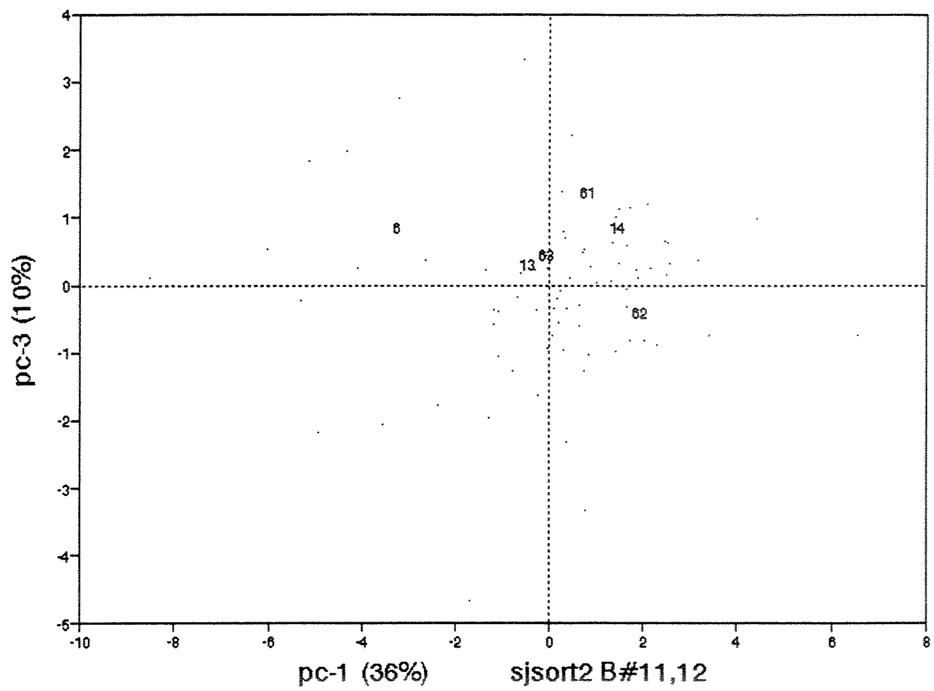

Fig.17 Projection of PAH profiles on the plane of the principal components 1 and 3 (pc-1 and pc-3). Numbers indicate samples from the Berth #11 and Berth #12 sites, samples from other sites are indicated by dots. Similar samples are located close together. Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

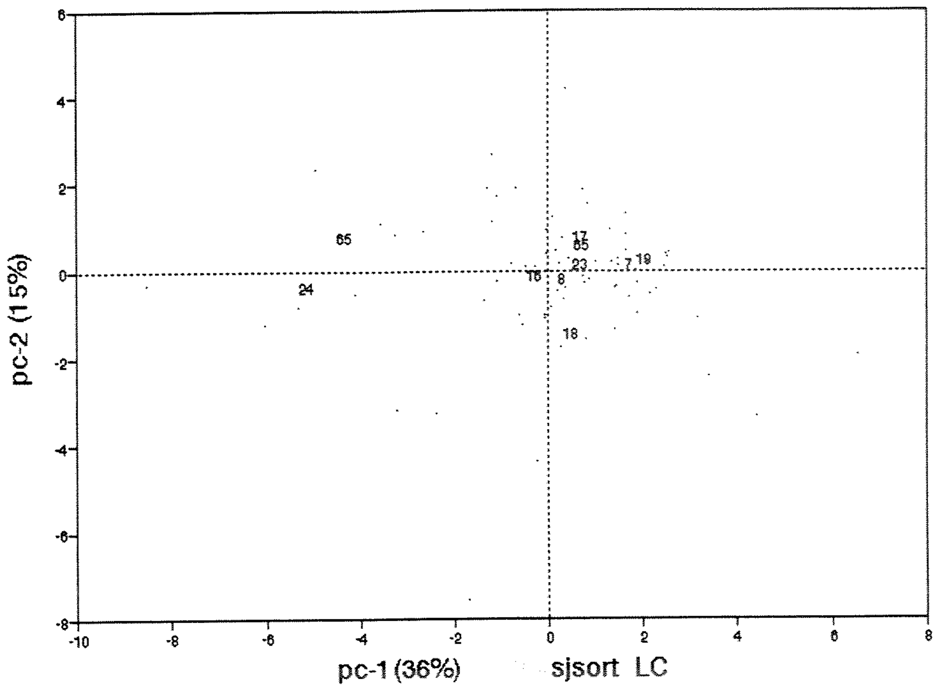

Fig.18 Projection of PAH profiles on the plane of the principal components 1 and 2 (pc-1 and pc-2). Numbers indicate samples from the Lower Cove site, samples from other sites are indicated by dots. Similar samples are located close together.
Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

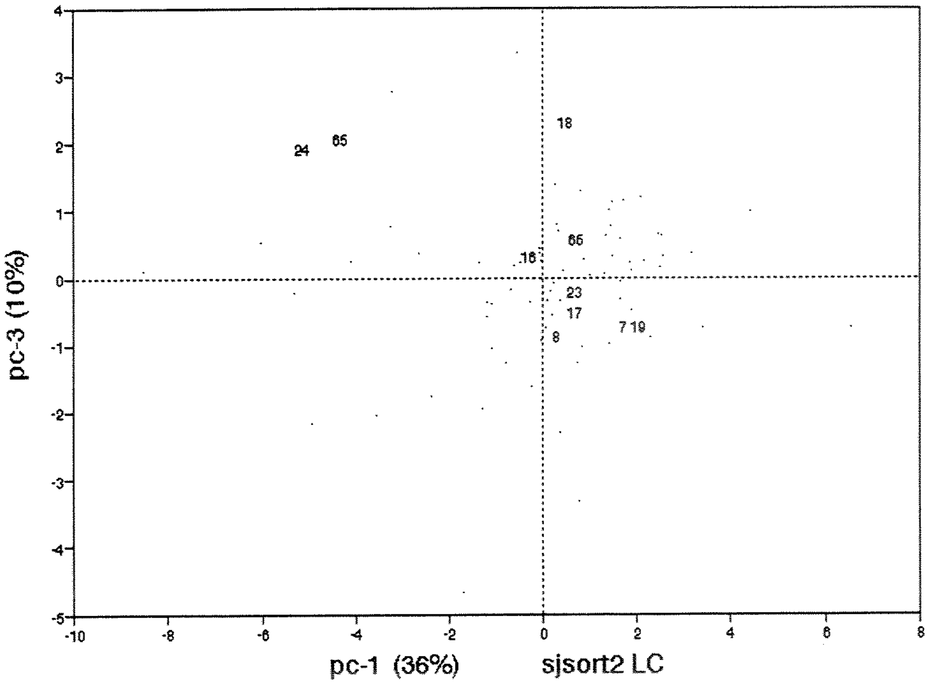

Fig.19 Projection of PAH profiles on the plane of the principal components 1 and 3 (pc-1 and pc-3). Numbers indicate samples from the Lower Cove site, samples from other sites are indicated by dots. Similar samples are located close together.
Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

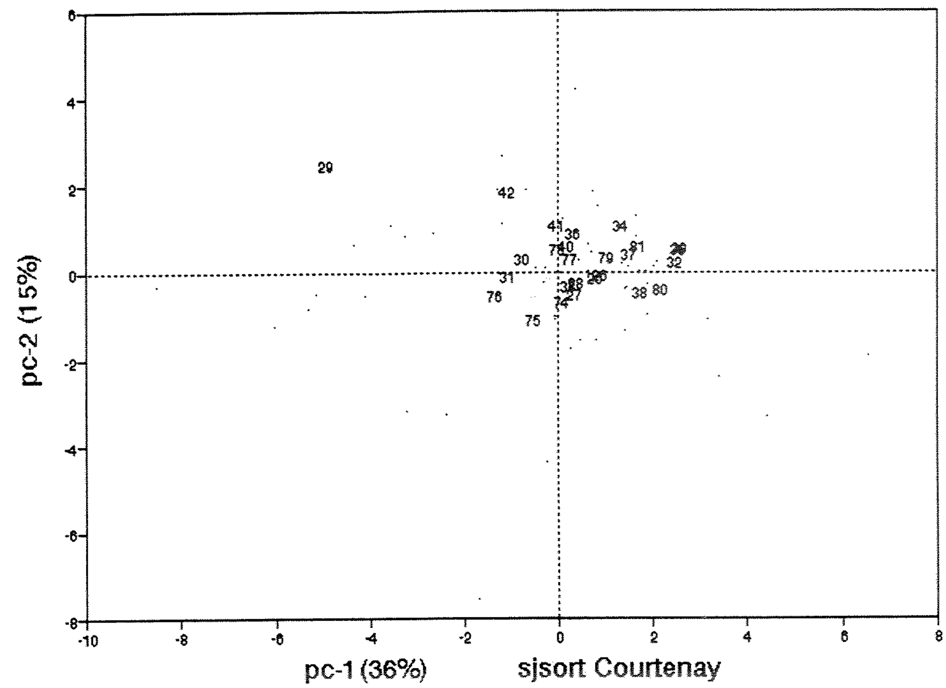

Fig.20 Projection of PAH profiles on the plane of the principal components 1 and 2 (pc-1 and pc-2). Numbers indicate samples from Courtenay Bay, samples from other sites are indicated by dots. Similar samples are located close together.
Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

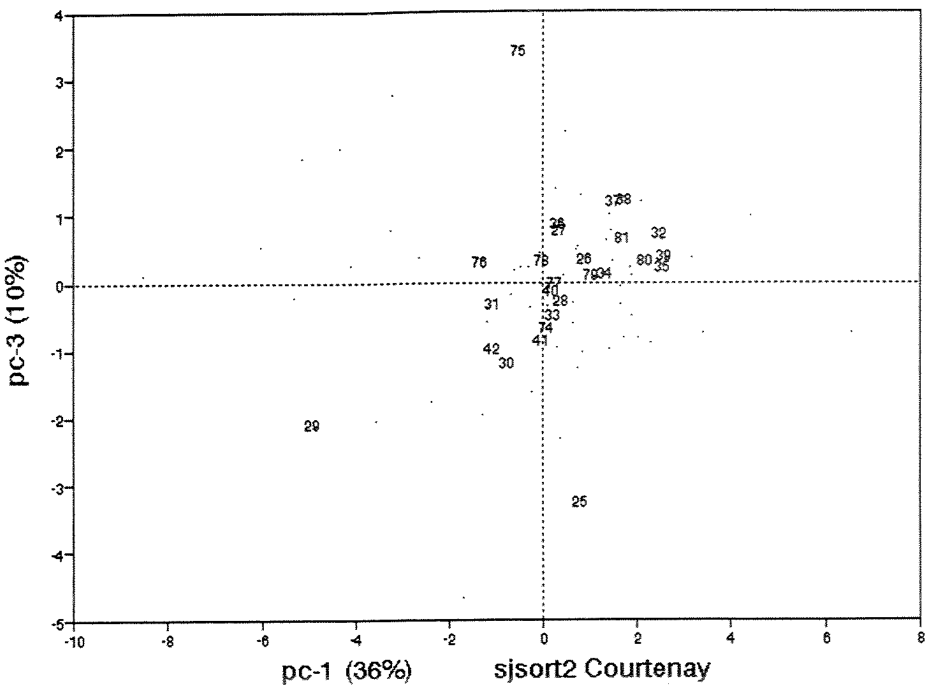

Fig.21 Projection of PAH profiles on the plane of the principal components 1 and 3 (pc-1 and pc-3). Numbers indicate samples from Courtenay Bay, samples from other sites are indicated by dots. Similar samples are located close together.
Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

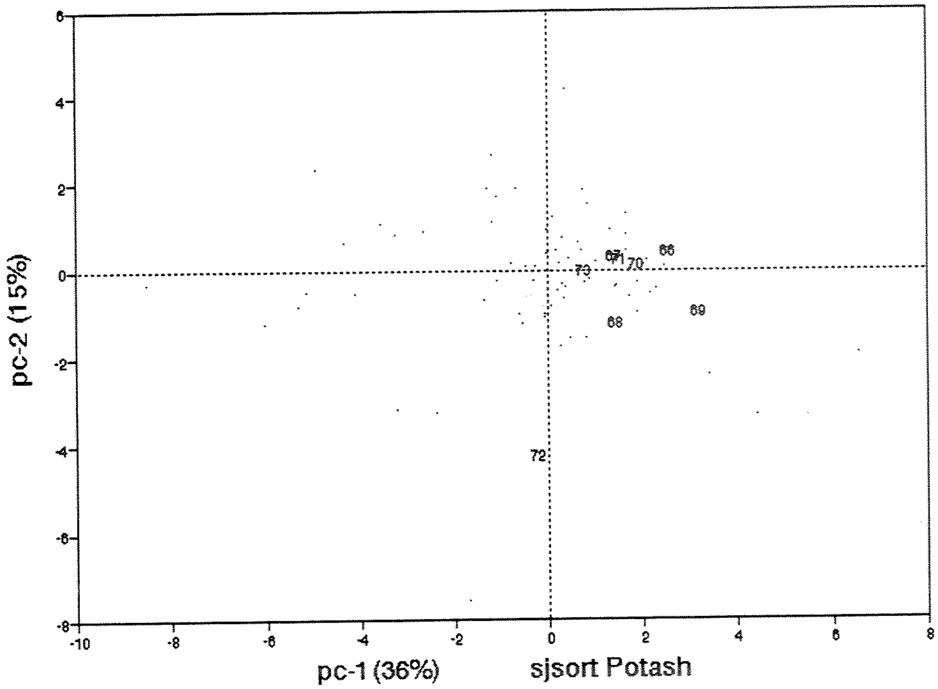

Fig.22 Projection of PAH profiles on the plane of the principal components 1 and 2 (pc-1 and pc-2). Numbers indicate samples from the Potash Terminal, samples from other sites are indicated by dots. Similar samples are located close together.
Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

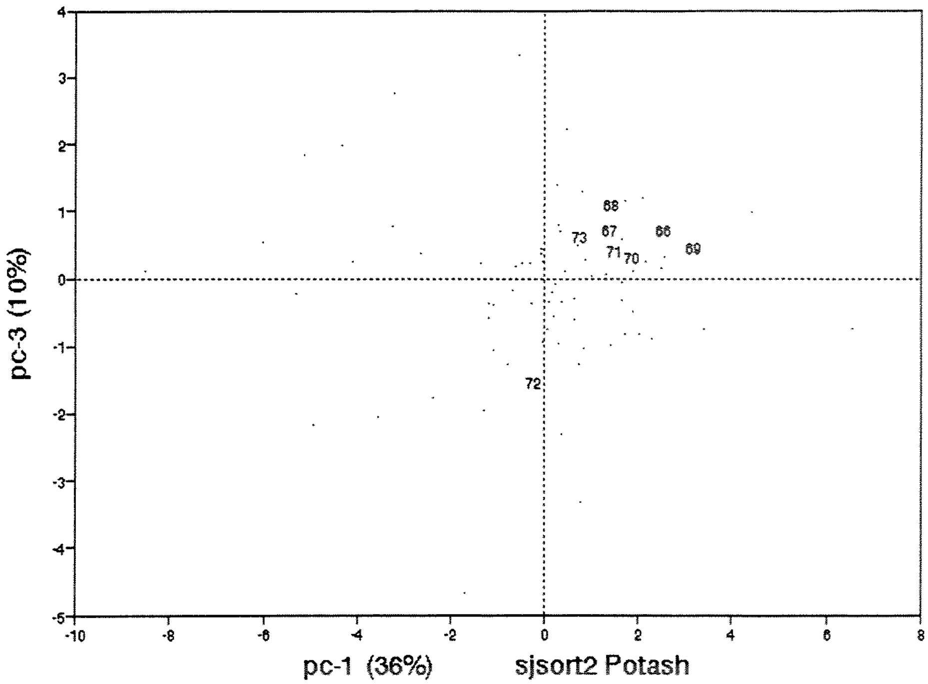

Fig.23 Projection of PAH profiles on the plane of the principal components 1 and 3 (pc-1 and pc-3). Numbers indicate samples from the Potash Terminal, samples from other sites are indicated by dots. Similar samples are located close together.
Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

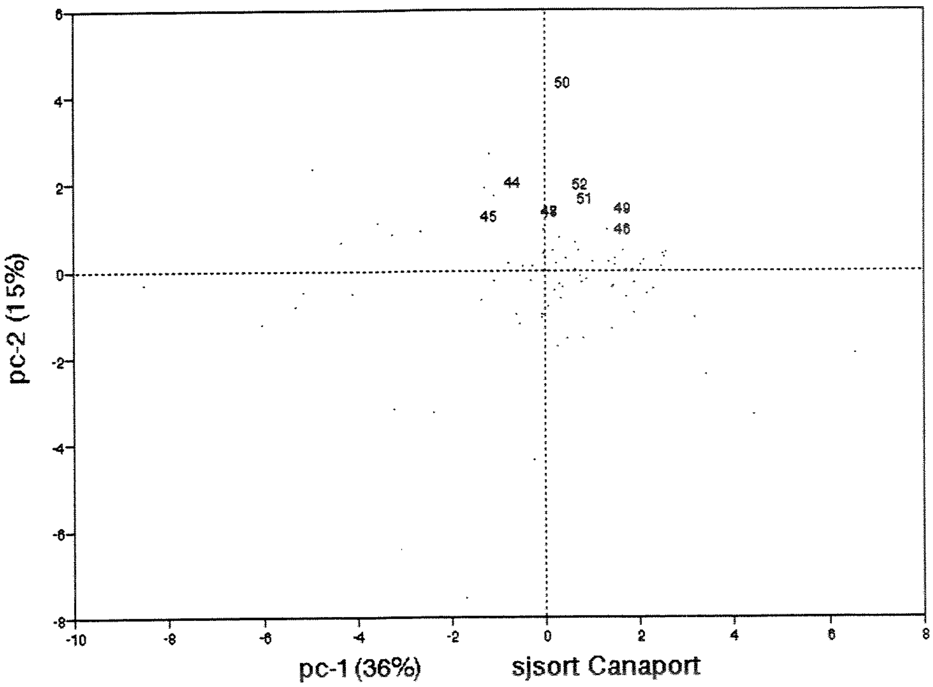

Fig.24 Projection of PAH profiles on the plane of the principal components 1 and 2 (pc-1 and pc-2). Numbers indicate samples from Canaport, samples from other sites are indicated by dots. Similar samples are located close together.
Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

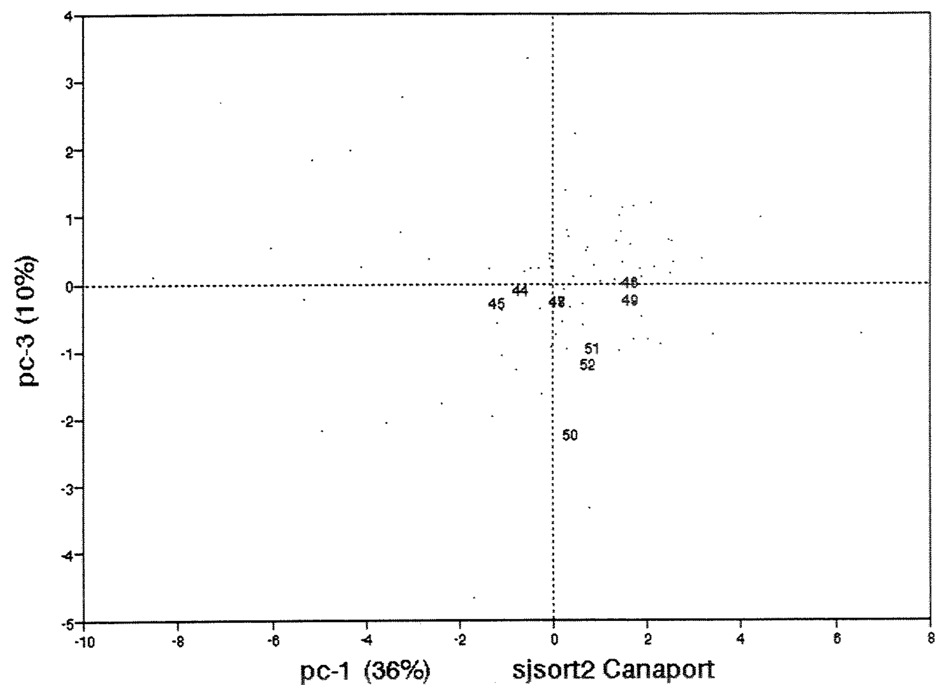

Fig.25 Projection of PAH profiles on the plane of the principal components 1 and 3 (pc-1 and pc-3). Numbers indicate samples from Canaport, samples from other sites are indicated by dots. Similar samples are located close together.
Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

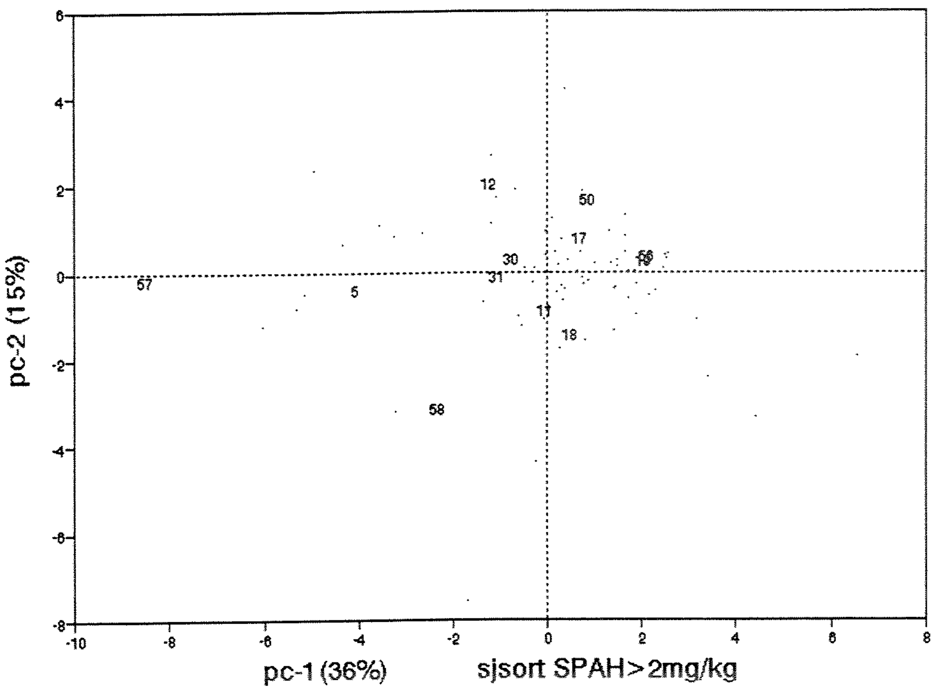

Fig.26 Projection of PAH profiles on the plane of the principal components 1 and 2 (pc-1 and pc-2). Numbers indicate samples with total PAH (SPAH) > 2.0 mg/kg, samples with lower SPAH are indicated by dots. Similar samples are located close together. Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

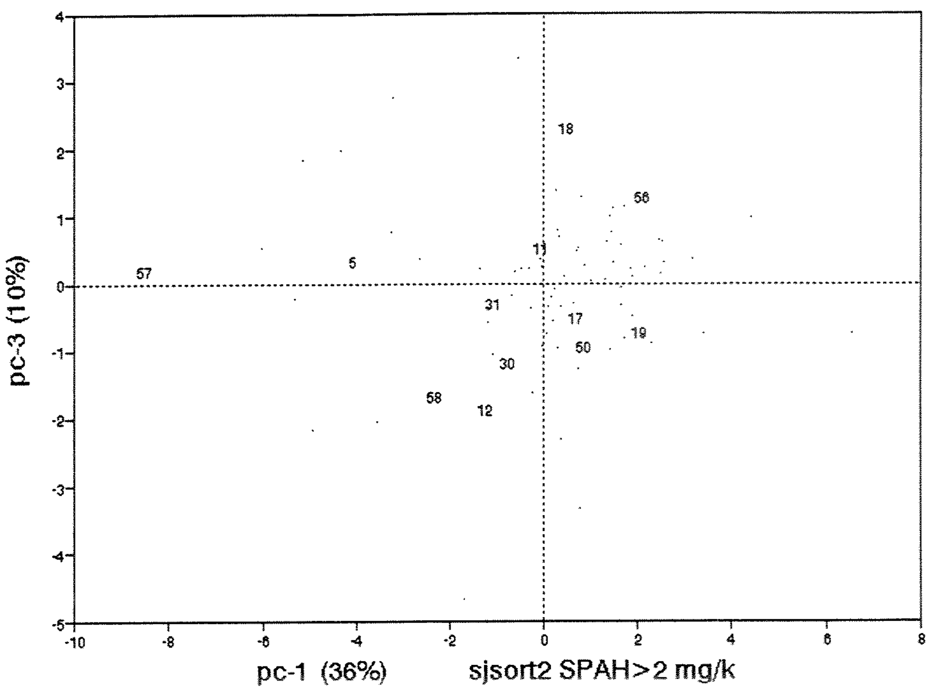

Fig.27 Projection of PAH profiles on the plane of the principal components 1 and 3 (pc-1 and pc-3). Numbers indicate samples with total PAH (SPAH) > 2.0 mg/kg, samples with lower SPAH are indicated by dots. Similar samples are located close together. Fractions of original variance accounted for by principal components are indicated in brackets. For example, pc-1 accounts for 36% of the original variance. For information on sampling stations see Appendices 1-4.

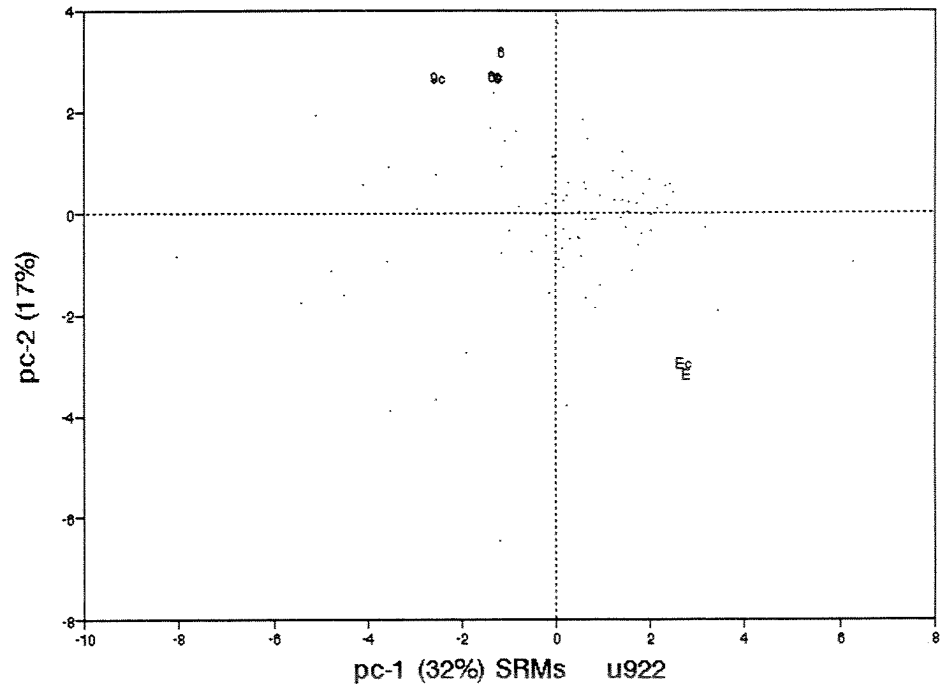

Fig.28 Profiles of the standard reference sediment HS-5 in 1996 (6 and 6c, reported and certified, respectively) and in 1999 (9 and 9c, reported and certified, respectively) and EC-2 (E and Ec, reported and certified, respectively), projected on the plane of the first two principal components. Note that the HS-5 'certified' profiles differ between 1996 and 1999 and that the profiles are quite different from those of the samples (dots).

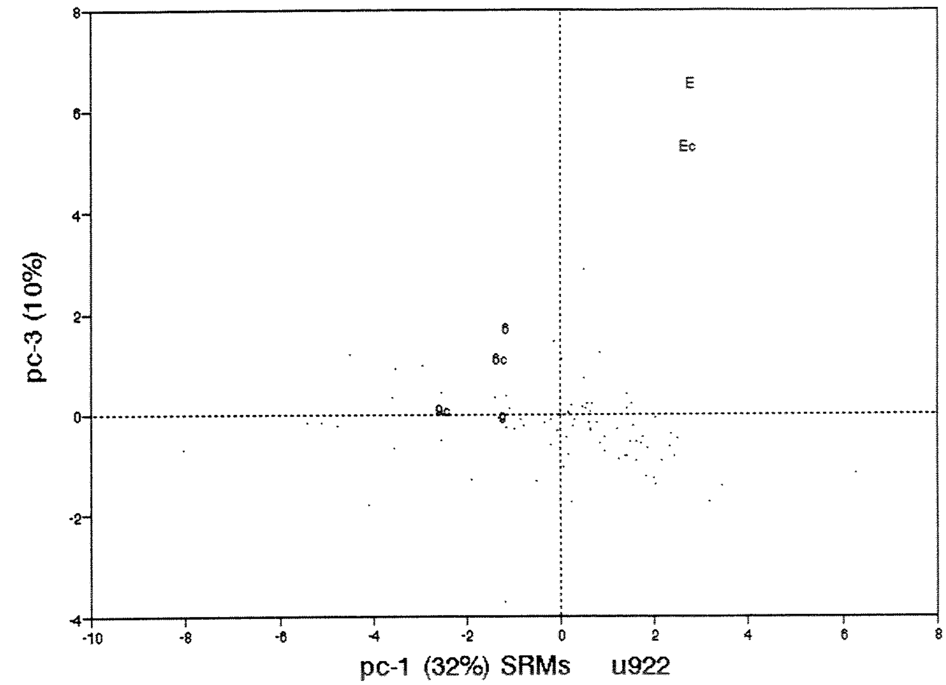

Fig.29 Profiles of the standard reference sediment HS-5 in 1996 (6 and 6c, reported and certified, respectively) and in 1999 (9 and 9c, reported and certified, respectively) and EC-2 (E and Ec, reported and certified, respectively), projected on the plane of principal components pc-1 and pc-3. Note that the HS-5 'certified' profiles differ between 1996 and 1999.

Appendix 1. Sediment samples from 1996, 1998, and 1999, with >0.5 mg/kg of total PAHs

#	Sample & site	Total PAHs (SPAH), mg/kg	Horwitz RSD Horwitz&Albert, 1997	Year
1	s12 Berth #3	0.71	108	96
2	s15 Rodney Slip	1.04	106	96
3	s16 Rodney Marginal	1.2	103	96
4	s19 Rodney Marginal	0.93	106	96
5	s20 Rodney Marginal	2.1	93	96
6	s21 Berth #11	1.01	103	96
7	s34 Lower Cove	1.185	99	96
8	s35 Lower Cove	1.155	99	96
9	s3 Berth #1	0.905	109	98
10	s8 Berth #3	0.94	105	98
11	s13 Rodney Marginal	3.34	84	98
12	s17 Rodney Marginal	6.225	82	98
13	s18 Berth #12	0.74	106	98
14	s20 Berth #12	0.525	111	98
15	s23 Long Wharf Slip	0.81	106	98
16	s24 LOWER COVE	1.73	94	98
17	s25 LOWER COVE	2.11	93	98
18	s26 LOWER COVE	4.46	80	98
19	s27 Lower Cove	3.62	86	98
20	s34 RODNEY SLIP	0.52	110	98
21	s35 RODNEY MARGINAL	1.495	97	98
22	s36 RODNEY MARGINAL	0.6	107	98
22a	S37 RODNEY MARGINAL	1.70	95	98
23	s42 LOWER COVE	1.065	101	98
24	s43 LOWER COVE	1.05	102	98
25	S44 COURTENAY BAY	1.405	98	98
26	s45 COURTENAY BAY	0.62	109	98
27	s46 COURTENAY BAY	0.73	106	98
28	s48 COURTENAY BAY	1.215	99	98
29	s49 COURTENAY BAY	1.575	104	98
30	s50 COURTENAY BAY	3.25	88	98
31	s50D COURTENAY BAY	2.38	91	98
32	s54 COURTENAY BAY	0.64	109	98
33	s55 COURTENAY BAY	1.085	101	98
34	s57 COURTENAY BAY	0.6	111	98
35	s58 COURTENAY BAY	0.65	109	98
36	s59 COURTENAY BAY	0.58	110	98
37	s60 COURTENAY BAY	0.5375	111	98
38	s61 COURTENAY BAY	0.57	110	98
39	s62 COURTENAY BAY	0.63	110	98
40	s63 COURTENAY BAY	0.825	105	98
41	s64 COURTENAY BAY	1.385	99	98

42	s65 COURTENAY BAY	1.465	99	98
43	s66 Canaport Box #04	0.685	111	98
44	s67 Canaport Box #10	0.6	112	98
45	s68 Canaport Box #12	0.765	106	98
46	s69 Canaport Box #13	0.685	107	98
47	s70 Canaport Box #19	0.615	110	98
48	s71 Canaport Box #25	0.615	110	98
49	s72 Canaport Box #36	0.675	108	98
50	s73 Canaport Box #39	8.77	80	98
51	s74 Canaport Box #43	1.165	102	98
52	s75 Canaport Box #48	1.545	100	98
53	s5 Berth #1	0.75	108	99
54	s11 Berth #3	0.65	108	99
55	s13 Berth #3	0.955	103	99
56	s15 RODNEY SLIP	4.75	79	99
57	s19 RODNEY MARGINAL	5.16	88	99
58	s20 RODNEY MARGINAL	2.245	93	99
59	s21 RODNEY MARGINAL	0.73	107	99
60	s25 RODNEY MARGINAL	1.195	100	99
61	s28 Berth #12	0.67	106	99
62	s29 Berth #12	1.215	99	99
63	s31 Berth #12	0.76	105	99
64	s37 LOWER COVE	0.535	111	99
65	s38 LOWER COVE	1.155	102	99
66	s39 POTASH TERMINAL	0.635	109	99
67	s40 POTASH TERMINAL	0.665	109	99
68	s41 POTASH TERMINAL	0.715	107	99
69	s42 POTASH TERMINAL	0.74	107	99
70	s43 POTASH TERMINAL	0.7	107	99
71	s44 POTASH TERMINAL	0.725	108	99
72	s45 POTASH TERMINAL	0.96	103	99
73	s46 POTASH TERMINAL	0.745	107	99
74	s49 COURTENAY BAY	1.195	99	99
75	50A COURTENAY BAY	0.625	109	99
76	s52 COURTENAY BAY	0.76	105	99
77	s53 COURTENAY BAY	1.03	101	99
78	55A COURTENAY BAY	0.565	111	99
79	s56 COURTENAY BAY	0.585	110	99
80	s57 COURTENAY BAY	0.57	110	99
81	s58 COURTENAY BAY	0.585	110	99

Appendix 2. Sediment samples from 1996, 1998, and 1999, with >0.5 mg/kg of total PAHs sorted according to locations

Table 1 number	Sample (original number, location)	Total PAHs (SPAH), mg/kg	Year
2	s15 Rodney Slip	1.04	96
20	s34 RODNEY SLIP	0.52	98
56	s15 RODNEY SLIP	4.75	99
3	s16 Rodney Marginal	1.2	96
4	s19 Rodney Marginal	0.93	96
5	s20 Rodney Marginal	2.1	96
11	s13 Rodney Marginal	3.34	98
12	s17 Rodney Marginal	6.225	98
21	s35 RODNEY MARGINAL	1.495	98
22	s36 RODNEY MARGINAL	0.6	98
22a	S37 Rodney Marginal	1.70	98
57	s19 RODNEY MARGINAL	5.16	99
58	s20 RODNEY MARGINAL	2.245	99
59	s21 RODNEY MARGINAL	0.73	99
60	s25 RODNEY MARGINAL	1.195	99
9	s3 Berth #1	0.905	98
53	s5 Berth #1	0.75	99
1	s12 Berth #3	0.71	96
10	s8 Berth #3	0.94	98
54	s11 Berth #3	0.65	99
55	s13 Berth #3	0.955	99
6	s21 Berth #11	1.01	96
13	s18 Berth #12	0.74	98
14	s20 Berth #12	0.525	98
61	s28 Berth #12	0.67	99
62	s29 Berth #12	1.215	99
63	s31 Berth #12	0.76	99
7	s34 Lower Cove	1.185	96
8	s35 Lower Cove	1.155	96
16	s24 LOWER COVE	1.73	98
17	s25 LOWER COVE	2.11	98
18	s26 LOWER COVE	4.46	98
19	s27 Lower Cove	3.62	98
23	s42 LOWER COVE	1.065	98
24	s43 LOWER COVE	1.05	98
64	s37 LOWER COVE	0.535	99
65	s38 LOWER COVE	1.155	99
15	s23 Long Wharf Slip	0.81	98
25	S44 COURTENAY BAY	1.405	98
26	s45 COURTENAY BAY	0.62	98
27	s46 COURTENAY BAY	0.73	98

28	s48 COURTENAY BAY	1.215	98
29	s49 COURTENAY BAY	1.575	98
30	s50 COURTENAY BAY	3.25	98
31	s50D COURTENAY BAY	2.38	98
32	s54 COURTENAY BAY	0.64	98
33	s55 COURTENAY BAY	1.085	98
34	s57 COURTENAY BAY	0.6	98
35	s58 COURTENAY BAY	0.65	98
36	s59 COURTENAY BAY	0.58	98
37	s60 COURTENAY BAY	0.5375	98
38	s61 COURTENAY BAY	0.57	98
39	s62 COURTENAY BAY	0.63	98
40	s63 COURTENAY BAY	0.825	98
41	s64 COURTENAY BAY	1.385	98
42	s65 COURTENAY BAY	1.465	98
74	s49 COURTENAY BAY	1.195	99
75	50A COURTENAY BAY	0.625	99
76	s52 COURTENAY BAY	0.76	99
77	s53 COURTENAY BAY	1.03	99
78	55A COURTENAY BAY	0.565	99
79	s56 COURTENAY BAY	0.585	99
80	s57 COURTENAY BAY	0.57	99
81	s58 COURTENAY BAY	0.585	99
66	s39 POTASH TERMINAL	0.635	99
67	s40 POTASH TERMINAL	0.665	99
68	s41 POTASH TERMINAL	0.715	99
69	s42 POTASH TERMINAL	0.74	99
70	s43 POTASH TERMINAL	0.7	99
71	s44 POTASH TERMINAL	0.725	99
72	s45 POTASH TERMINAL	0.96	99
73	s46 POTASH TERMINAL	0.745	99
43	s66 Canaport Box #04	0.685	98
44	s67 Canaport Box #10	0.6	98
45	s68 Canaport Box #12	0.765	98
46	s69 Canaport Box #13	0.685	98
47	s70 Canaport Box #19	0.615	98
48	s71 Canaport Box #25	0.615	98
49	s72 Canaport Box #36	0.675	98
50	s73 Canaport Box #39	8.77	98
51	s74 Canaport Box #43	1.165	98
52	s75 Canaport Box #48	1.545	98

Appendix 3. Concentration of PAHs in mg/kg dry sediment

#	Site	N	Acy	Ace	Fen	Phe	Ant	Flu	Pyr	BaA	Chr	BFl	BaP	ind	DahA	ghi	SPAH
1	s12 3	0.01	0.005	0.01	0.02	0.1	0.03	0.13	0.1	0.05	0.06	0.07	0.04	0.03	0.005	0.02	0.68
2	s15 rs	0.01	0.005	0.01	0.02	0.16	0.1	0.25	0.19	0.04	0.07	0.07	0.03	0.03	0.005	0.02	1.01
3	s16 rm	0.01	0.005	0.06	0.06	0.26	0.04	0.24	0.19	0.06	0.07	0.08	0.04	0.03	0.005	0.02	1.17
4	s19 rm	0.01	0.005	0.06	0.05	0.2	0.03	0.2	0.14	0.04	0.04	0.06	0.03	0.02	0.005	0.02	0.91
5	s20 rm	0.06	0.01	0.08	0.1	0.37	0.08	0.44	0.32	0.12	0.13	0.15	0.09	0.04	0.01	0.04	2.04
6	s21 11	0.05	0.005	0.04	0.03	0.11	0.04	0.25	0.16	0.05	0.06	0.09	0.04	0.02	0.005	0.02	0.97
7	s34 lc	0.03	0.005	0.02	0.02	0.14	0.05	0.18	0.16	0.1	0.11	0.12	0.08	0.05	0.02	0.05	1.14
8	s35 lc	0.03	0.005	0.02	0.02	0.17	0.06	0.18	0.16	0.08	0.09	0.11	0.07	0.05	0.02	0.04	1.11
9	98 s3 #1	0.01	0.005	0.005	0.03	0.1	0.39	0.06	0.05	0.03	0.05	0.06	0.03	0.03	0.005	0.02	0.88
10	s8 #3	0.01	0.005	0.01	0.02	0.13	0.05	0.14	0.11	0.07	0.09	0.12	0.06	0.04	0.005	0.03	0.89
11	s13 rm	0.14	0.02	0.08	0.1	0.48	0.16	0.48	0.43	0.23	0.28	0.35	0.22	0.12	0.02	0.09	3.20
12	s17 rm	0.11	0.005	0.08	0.11	0.68	0.33	1.34	1.18	0.43	0.48	0.58	0.3	0.21	0.04	0.14	6.02
13	s18 rm	0.02	0.005	0.02	0.02	0.09	0.03	0.13	0.11	0.05	0.06	0.08	0.04	0.03	0.005	0.02	0.71
14	s20 #12	0.015	0.005	0.01	0.01	0.055	0.02	0.085	0.065	0.03	0.045	0.07	0.03	0.03	0.005	0.02	0.50
15	s23 Lwslip	0.02	0.005	0.01	0.02	0.08	0.03	0.12	0.11	0.05	0.06	0.11	0.07	0.04	0.005	0.03	0.76
16	s24 LC	0.06	0.01	0.03	0.04	0.23	0.07	0.3	0.26	0.09	0.12	0.18	0.1	0.08	0.01	0.07	1.65
17	s25 LC	0.05	0.01	0.02	0.04	0.24	0.09	0.37	0.33	0.14	0.17	0.24	0.14	0.09	0.01	0.07	2.01
18	s26 LC	0.43	0.03	0.06	0.12	0.55	0.16	0.65	0.53	0.27	0.33	0.45	0.3	0.19	0.04	0.15	4.26
19	s27 Lc	0.06	0.02	0.02	0.06	0.4	0.19	0.58	0.48	0.29	0.32	0.44	0.24	0.18	0.03	0.14	3.45
20	s34 RS	0.02	0.005	0.01	0.02	0.07	0.02	0.08	0.06	0.03	0.04	0.06	0.03	0.03	0.005	0.02	0.50
21	s35 RM	0.08	0.005	0.02	0.03	0.17	0.05	0.23	0.24	0.1	0.12	0.17	0.09	0.06	0.01	0.05	1.43
22	s36 RM	0.04	0.005	0.04	0.03	0.1	0.03	0.08	0.07	0.03	0.04	0.05	0.02	0.02	0.005	0.02	0.58
22a	s37 RM	0.08	0.005	0.1	0.12	0.32	0.1	0.26	0.18	0.09	0.11	0.14	0.07	0.04	0.01	0.03	1.66
23	s42 LC	0.03	0.005	0.02	0.02	0.14	0.04	0.17	0.15	0.07	0.08	0.13	0.07	0.04	0.01	0.04	1.02
24	s43 LC	0.05	0.005	0.09	0.05	0.12	0.04	0.22	0.18	0.05	0.05	0.08	0.04	0.02	0.005	0.02	1.02
25	S44 C	0.02	0.005	0.01	0.03	0.2	0.07	0.22	0.18	0.08	0.1	0.15	0.09	0.05	0.1	0.04	1.35
26	s45 C	0.02	0.005	0.01	0.01	0.08	0.03	0.09	0.09	0.04	0.05	0.07	0.04	0.03	0.005	0.02	0.59
27	s46 C	0.03	0.005	0.01	0.02	0.09	0.03	0.12	0.1	0.04	0.05	0.08	0.05	0.03	0.005	0.03	0.69
28	s48 C	0.04	0.005	0.02	0.03	0.15	0.06	0.19	0.17	0.08	0.09	0.14	0.08	0.05	0.01	0.04	1.16
29	s49 C	0.01	0.005	0.01	0.02	0.38	0.03	0.42	0.31	0.04	0.1	0.1	0.04	0.03	0.01	0.03	1.54
30	s50 C	0.06	0.01	0.04	0.07	0.53	0.16	0.56	0.5	0.2	0.24	0.33	0.19	0.11	0.02	0.1	3.12
31	s50D C	0.07	0.01	0.04	0.06	0.36	0.12	0.41	0.37	0.14	0.16	0.22	0.13	0.1	0.01	0.08	2.28
32	s54 C	0.02	0.005	0.01	0.01	0.06	0.02	0.1	0.08	0.05	0.06	0.08	0.04	0.03	0.005	0.03	0.60
33	s55 C	0.02	0.005	0.02	0.03	0.15	0.05	0.17	0.15	0.07	0.08	0.13	0.07	0.04	0.01	0.04	1.04
34	s57 C	0.01	0.005	0.01	0.01	0.06	0.02	0.1	0.09	0.04	0.05	0.08	0.04	0.02	0.005	0.02	0.56
35	s58 C	0.01	0.005	0.01	0.01	0.06	0.02	0.11	0.08	0.05	0.06	0.09	0.04	0.03	0.005	0.03	0.61
36	s59 C	0.02	0.005	0.01	0.01	0.06	0.02	0.12	0.08	0.04	0.04	0.07	0.03	0.02	0.005	0.02	0.55
37	s60 C	0.02	0.005	0.0075	0.01	0.05	0.015	0.095	0.07	0.035	0.04	0.08	0.03	0.025	0.005	0.02	0.51
38	s61 C	0.02	0.005	0.01	0.01	0.06	0.02	0.09	0.07	0.03	0.04	0.09	0.03	0.03	0.005	0.03	0.54
39	s62 C	0.01	0.005	0.01	0.01	0.05	0.02	0.11	0.08	0.04	0.05	0.11	0.04	0.03	0.005	0.03	0.60
40	s63 C	0.02	0.005	0.01	0.02	0.09	0.03	0.17	0.11	0.05	0.06	0.11	0.04	0.03	0.01	0.03	0.79
41	s64 C	0.02	0.005	0.02	0.03	0.16	0.05	0.28	0.2	0.09	0.1	0.2	0.07	0.05	0.01	0.04	1.33
42	s65 C	0.02	0.005	0.02	0.03	0.17	0.05	0.34	0.24	0.09	0.09	0.19	0.07	0.04	0.01	0.04	1.41
43	s66 C	0.01	0.005	0.005	0.01	0.07	0.02	0.19	0.12	0.04	0.05	0.07	0.03	0.02	0.005	0.02	0.67
44	s67 C	0.01	0.005	0.01	0.01	0.07	0.02	0.14	0.1	0.04	0.05	0.06	0.03	0.01	0.005	0.02	0.58
45	s68 C	0.02	0.005	0.01	0.02	0.09	0.03	0.17	0.12	0.05	0.06	0.08	0.03	0.02	0.01	0.02	0.74
46	s69 C	0.02	0.005	0.01	0.01	0.07	0.02	0.13	0.09	0.05	0.07	0.08	0.04	0.02	0.01	0.03	0.66
47	s70 C	0.01	0.005	0.01	0.01	0.07	0.02	0.13	0.09	0.04	0.05	0.07	0.03	0.02	0.01	0.02	0.59
48	s71 C	0.01	0.005	0.01	0.01	0.07	0.02	0.13	0.09	0.04	0.05	0.07	0.03	0.02	0.01	0.02	0.59
49	s72 C	0.01	0.005	0.01	0.01	0.06	0.02	0.13	0.1	0.05	0.06	0.09	0.04	0.02	0.01	0.03	0.65
50	s73 C	0.05	0.01	0.06	0.11	0.41	0.23	1.83	2.11	0.67	0.78	1.11	0.55	0.19	0.07	0.2	8.38
51	s74 C	0.02	0.005	0.01	0.02	0.12	0.04	0.24	0.17	0.09	0.11	0.15	0.06	0.03	0.01	0.04	1.12
52	s75 C	0.02	0.005	0.01	0.03	0.16	0.04	0.32	0.24	0.13	0.15	0.19	0.08	0.04	0.01	0.05	1.48
53	s5 99 #1	0.005	0.005	0.005	0.02	0.09	0.07	0.08	0.08	0.05	0.07	0.11	0.06	0.05	0.005	0.05	0.75
54	s11 99	0.01	0.005	0.01	0.02	0.12	0.03	0.11	0.09	0.05	0.04	0.07	0.03	0.03	0.005	0.03	0.65
55	s13 99 #3	0.01	0.005	0.01	0.02	0.13	0.04	0.14	0.12	0.09	0.1	0.12	0.06	0.06	0.01	0.04	0.96
56	s15 99 RS	0.09	0.06	0.09	0.1	0.47	0.13	0.87	0.61	0.47	0.44	0.65	0.24	0.29	0.06	0.18	4.75

#	Site	N	Acy	Ace	Fen	Phe	Ant	Flu	Pyr	BaA	Chr	BFl	BaP	ind	DahA	ghi	SPAH
57	s19 99 RM	0.03	0.02	0.36	0.25	1.5	0.07	1.2	0.96	0.26	0.14	0.17	0.08	0.07	0.01	0.04	5.16
58	s20 99 RM	0.04	0.005	0.02	0.11	0.47	0.3	0.31	0.26	0.16	0.15	0.16	0.09	0.09	0.01	0.07	2.25
59	s21 99 RM	0.01	0.005	0.005	0.01	0.07	0.04	0.06	0.05	0.07	0.11	0.12	0.06	0.07	0.01	0.04	0.73
60	s25 99 RM	0.04	0.01	0.03	0.03	0.19	0.05	0.25	0.22	0.08	0.09	0.1	0.04	0.04	0.005	0.02	1.20
61	s28 99 #1	0.03	0.005	0.01	0.02	0.1	0.03	0.11	0.08	0.06	0.04	0.07	0.04	0.03	0.005	0.04	0.67
62	s29 99 #1	0.03	0.005	0.02	0.02	0.16	0.06	0.18	0.15	0.11	0.11	0.13	0.07	0.11	0.01	0.05	1.22
63	s31 99 #1	0.02	0.005	0.02	0.02	0.12	0.04	0.12	0.1	0.06	0.05	0.09	0.04	0.04	0.005	0.03	0.76
64	s37 99 LC	0.02	0.005	0.005	0.01	0.07	0.02	0.1	0.08	0.04	0.05	0.06	0.03	0.02	0.005	0.02	0.54
65	s38 99 LC	0.02	0.02	0.02	0.04	0.26	0.02	0.28	0.21	0.06	0.05	0.08	0.04	0.03	0.005	0.02	1.16
66	s39 99 P	0.02	0.005	0.005	0.01	0.06	0.02	0.11	0.09	0.06	0.05	0.09	0.04	0.04	0.005	0.03	0.64
67	s40 99 P	0.02	0.005	0.005	0.01	0.08	0.02	0.13	0.1	0.05	0.04	0.08	0.04	0.04	0.005	0.04	0.67
68	s41 99 P	0.03	0.005	0.005	0.02	0.09	0.02	0.12	0.09	0.04	0.05	0.1	0.04	0.06	0.005	0.04	0.72
69	s42 99 P	0.02	0.005	0.005	0.01	0.07	0.02	0.12	0.09	0.06	0.05	0.09	0.05	0.11	0.01	0.03	0.74
70	s43 99 P	0.02	0.005	0.005	0.01	0.08	0.02	0.13	0.1	0.05	0.05	0.09	0.04	0.05	0.01	0.04	0.70
71	s44 99 P	0.02	0.005	0.005	0.01	0.08	0.02	0.14	0.11	0.04	0.05	0.1	0.04	0.06	0.005	0.04	0.73
72	s45 99 P	0.02	0.005	0.005	0.04	0.11	0.2	0.12	0.1	0.05	0.05	0.1	0.04	0.06	0.01	0.05	0.96
73	s46 99 P	0.03	0.005	0.005	0.02	0.09	0.03	0.14	0.11	0.06	0.06	0.08	0.04	0.03	0.005	0.04	0.75
74	s49 99 C	0.03	0.005	0.02	0.03	0.2	0.06	0.17	0.18	0.08	0.1	0.12	0.08	0.07	0.01	0.04	1.20
75	50A99 C	0.08	0.005	0.005	0.02	0.08	0.02	0.1	0.09	0.05	0.04	0.06	0.03	0.02	0.005	0.02	0.63
76	s52 99 C	0.03	0.005	0.02	0.02	0.12	0.04	0.14	0.11	0.04	0.06	0.07	0.04	0.04	0.005	0.02	0.76
77	s53 99 C	0.02	0.01	0.01	0.02	0.15	0.05	0.19	0.16	0.06	0.09	0.11	0.06	0.05	0.01	0.04	1.03
78	55A99 C	0.02	0.005	0.005	0.01	0.08	0.02	0.11	0.09	0.03	0.05	0.06	0.03	0.03	0.005	0.02	0.57
79	s56 99 C	0.02	0.005	0.005	0.01	0.07	0.03	0.11	0.08	0.04	0.06	0.07	0.03	0.03	0.005	0.02	0.59
80	s57 99 C	0.01	0.005	0.005	0.01	0.07	0.02	0.09	0.08	0.05	0.04	0.07	0.03	0.05	0.01	0.03	0.57
81	s58 99 C	0.02	0.005	0.005	0.01	0.05	0.02	0.11	0.09	0.05	0.05	0.07	0.03	0.05	0.005	0.02	0.59

Appendix 4. Concentration of PAHs in %

#	Site	N	Acy	Ace	Fen	Phe	Ant	Flu	Pyr	BaA	Chr	BFl	BaP	ind	DahA	ghi
1	s3 12	1.47	0.74	1.47	2.94	14.71	4.41	19.12	14.71	7.35	8.82	10.29	5.88	4.41	0.74	2.94
2	s15 rs	0.99	0.50	0.99	1.98	15.84	9.90	24.75	18.81	3.96	6.93	6.93	2.97	2.97	0.50	1.98
3	s16 rm	0.85	0.43	5.13	5.13	22.22	3.42	20.51	16.24	5.13	5.98	6.84	3.42	2.56	0.43	1.71
4	s19 rm	1.10	0.55	6.59	5.49	21.98	3.30	21.98	15.39	4.40	4.40	6.59	3.30	2.20	0.55	2.20
5	s20 rm	2.94	0.49	3.92	4.90	18.14	3.92	21.57	15.69	5.88	6.37	7.35	4.41	1.96	0.49	1.96
6	s21 11	5.15	0.52	4.12	3.09	11.34	4.12	25.77	16.50	5.15	6.19	9.28	4.12	2.06	0.52	2.06
7	s34 lc	2.64	0.44	1.76	1.76	12.34	4.41	15.86	14.10	8.81	9.69	10.57	7.05	4.41	1.76	4.41
8	s35 lc	2.71	0.45	1.81	1.81	15.39	5.43	16.29	14.48	7.24	8.14	9.95	6.33	4.52	1.81	3.62
9	98 s3 #1	1.14	0.57	0.57	3.43	11.43	44.57	6.86	5.71	3.43	5.71	6.86	3.43	3.43	0.57	2.29
10	s8 #3	1.12	0.56	1.12	2.25	14.61	5.62	15.73	12.36	7.87	10.11	13.48	6.74	4.49	0.56	3.37
11	s13 rm	4.38	0.63	2.50	3.13	15.00	5.00	15.00	13.44	7.19	8.75	10.94	6.88	3.75	0.63	2.81
12	s17 rm	1.83	0.08	1.33	1.83	11.31	5.49	22.28	19.62	7.15	7.98	9.64	4.99	3.49	0.67	2.33
13	s18 rm	2.82	0.70	2.82	2.82	12.68	4.23	18.31	15.49	7.04	8.45	11.27	5.63	4.23	0.70	2.82
14	s20 #12	3.03	1.01	2.02	2.02	11.11	4.04	17.17	13.13	6.06	9.09	14.14	6.06	6.06	1.01	4.04
15	s23 Lwslip	2.63	0.66	1.32	2.63	10.53	3.95	15.79	14.47	6.58	7.89	14.47	9.21	5.26	0.66	3.95
16	s24 LC	3.64	0.61	1.82	2.42	13.94	4.24	18.18	15.76	5.45	7.27	10.91	6.06	4.85	0.61	4.24
17	s25 LC	2.49	0.50	1.00	1.99	11.94	4.48	18.41	16.42	6.97	8.46	11.94	6.97	4.48	0.50	3.48
18	s26 LC	10.09	0.70	1.41	2.82	12.91	3.76	15.26	12.44	6.34	7.75	10.56	7.04	4.46	0.94	3.52
19	s27 Lc	1.74	0.58	0.58	1.74	11.59	5.51	16.81	13.91	8.41	9.28	12.75	6.96	5.22	0.87	4.06
20	s34 RS	4.00	1.00	2.00	4.00	14.00	4.00	16.00	12.00	6.00	8.00	12.00	6.00	6.00	1.00	4.00
21	s35 RM	5.61	0.35	1.40	2.11	11.93	3.51	16.14	16.84	7.02	8.42	11.93	6.32	4.21	0.70	3.51
22	s36 RM	6.90	0.86	6.90	5.17	17.24	5.17	13.79	12.07	5.17	6.90	8.62	3.45	3.45	0.86	3.45
22a	s37 RM	4.80	0.30	6.04	7.25	19.33	6.04	15.71	10.88	5.44	6.65	8.46	4.23	2.42	0.60	1.82
23	s42 LC	2.96	0.49	1.97	1.97	13.79	3.94	16.75	14.78	6.90	7.88	12.81	6.90	3.94	0.99	3.94
24	s43 LC	4.90	0.49	8.82	4.90	11.77	3.92	21.57	17.65	4.90	4.90	7.84	3.92	1.96	0.49	1.96
25	S44 C	1.49	0.37	0.74	2.23	14.87	5.20	16.36	13.38	5.95	7.43	11.15	6.69	3.72	7.43	2.97
26	s45 C	3.39	0.85	1.69	1.69	13.56	5.08	15.25	15.25	6.78	8.47	11.86	6.78	5.08	0.85	3.39
27	s46 C	4.35	0.72	1.45	2.90	13.04	4.35	17.39	14.49	5.80	7.25	11.59	7.25	4.35	0.72	4.35
28	s48 C	3.46	0.43	1.73	2.60	12.99	5.19	16.45	14.72	6.93	7.79	12.12	6.93	4.33	0.87	3.46
29	s49 C	0.65	0.33	0.65	1.30	24.76	1.95	27.36	20.20	2.61	6.51	6.51	2.61	1.95	0.65	1.95
30	s50 C	1.92	0.32	1.28	2.24	16.99	5.13	17.95	16.03	6.41	7.69	10.58	6.09	3.53	0.64	3.21
31	s50D C	3.07	0.44	1.75	2.63	15.79	5.26	17.98	16.23	6.14	7.02	9.65	5.70	4.39	0.44	3.51
32	s54 C	3.33	0.83	1.67	1.67	10.00	3.33	16.67	13.33	8.33	10.00	13.33	6.67	5.00	0.83	5.00
33	s55 C	1.93	0.48	1.93	2.90	14.49	4.83	16.43	14.49	6.76	7.73	12.56	6.76	3.86	0.97	3.86
34	s57 C	1.79	0.89	1.79	1.79	10.71	3.57	17.86	16.07	7.14	8.93	14.29	7.14	3.57	0.89	3.57
35	s58 C	1.64	0.82	1.64	1.64	9.84	3.28	18.03	13.12	8.20	9.84	14.75	6.56	4.92	0.82	4.92
36	s59 C	3.64	0.91	1.82	1.82	10.91	3.64	21.82	14.55	7.27	7.27	12.73	5.45	3.64	0.91	3.64
37	s60 C	3.94	0.99	1.48	1.97	9.85	2.96	18.72	13.79	6.90	7.88	15.76	5.91	4.93	0.99	3.94
38	s61 C	3.70	0.93	1.85	1.85	11.11	3.70	16.67	12.96	5.56	7.41	16.67	5.56	5.56	0.93	5.56
39	s62 C	1.67	0.83	1.67	1.67	8.33	3.33	18.33	13.33	6.67	8.33	18.33	6.67	5.00	0.83	5.00
40	s63 C	2.55	0.64	1.27	2.55	11.47	3.82	21.66	14.01	6.37	7.64	14.01	5.10	3.82	1.27	3.82
41	s64 C	1.51	0.38	1.51	2.26	12.08	3.77	21.13	15.09	6.79	7.55	15.09	5.28	3.77	0.75	3.02
42	s65 C	1.42	0.36	1.42	2.14	12.10	3.56	24.20	17.08	6.41	6.41	13.52	4.98	2.85	0.71	2.85
43	s66 C	1.50	0.75	0.75	1.50	10.53	3.01	28.57	18.05	6.02	7.52	10.53	4.51	3.01	0.75	3.01
44	s67 C	1.72	0.86	1.72	1.72	12.07	3.45	24.14	17.24	6.90	8.62	10.35	5.17	1.72	0.86	3.45
45	s68 C	2.72	0.68	1.36	2.72	12.25	4.08	23.13	16.33	6.80	8.16	10.88	4.08	2.72	1.36	2.72
46	s69 C	3.05	0.76	1.53	1.53	10.69	3.05	19.85	13.74	7.63	10.69	12.21	6.11	3.05	1.53	4.58
47	s70 C	1.71	0.85	1.71	1.71	11.97	3.42	22.22	15.39	6.84	8.55	11.97	5.13	3.42	1.71	3.42
48	s71 C	1.71	0.85	1.71	1.71	11.97	3.42	22.22	15.39	6.84	8.55	11.97	5.13	3.42	1.71	3.42
49	s72 C	1.55	0.78	1.55	1.55	9.30	3.10	20.16	15.50	7.75	9.30	13.95	6.20	3.10	1.55	4.65
50	s73 C	0.60	0.12	0.72	1.31	4.89	2.74	21.84	25.18	8.00	9.31	13.25	6.56	2.27	0.84	2.39
51	s74 C	1.79	0.45	0.90	1.79	10.76	3.59	21.53	15.25	8.07	9.87	13.45	5.38	2.69	0.90	3.59
52	s75 C	1.36	0.34	0.68	2.03	10.85	2.71	21.70	16.27	8.81	10.17	12.88	5.42	2.71	0.68	3.39
53	s5 99 #1	0.67	0.67	0.67	2.67	12.00	9.33	10.67	10.67	6.67	9.33	14.67	8.00	6.67	0.67	6.67
54	s11 99	1.54	0.77	1.54	3.08	18.46	4.62	16.92	13.85	7.69	6.15	10.77	4.62	4.62	0.77	4.62
55	s13 99 #3	1.05	0.52	1.05	2.09	13.61	4.19	14.66	12.57	9.42	10.47	12.57	6.28	6.28	1.05	4.19
56	s15 99 RS	1.89	1.26	1.89	2.11	9.89	2.74	18.32	12.84	9.89	9.26	13.68	5.05	6.11	1.26	3.79

#	Site	N	Acy	Ace	Fen	Phe	Ant	Flu	Pyr	BaA	Chr	BFl	BaP	ind	DahA	ghi
57	s19 99 RM	0.58	0.39	6.98	4.85	29.07	1.36	23.26	18.61	5.04	2.71	3.29	1.55	1.36	0.19	0.78
58	s20 99 RM	1.78	0.22	0.89	4.90	20.94	13.36	13.81	11.58	7.13	6.68	7.13	4.01	4.01	0.45	3.12
59	s21 99 RM	1.37	0.68	0.68	1.37	9.59	5.48	8.22	6.85	9.59	15.07	16.44	8.22	9.59	1.37	5.48
60	s25 99 RM	3.35	0.84	2.51	2.51	15.90	4.18	20.92	18.41	6.69	7.53	8.37	3.35	3.35	0.42	1.67
61	s28 99 #1	4.48	0.75	1.49	2.99	14.93	4.48	16.42	11.94	8.96	5.97	10.45	5.97	4.48	0.75	5.97
62	s29 99 #1	2.47	0.41	1.65	1.65	13.17	4.94	14.82	12.35	9.05	9.05	10.70	5.76	9.05	0.82	4.12
63	s31 99 #1	2.63	0.66	2.63	2.63	15.79	5.26	15.79	13.16	7.89	6.58	11.84	5.26	5.26	0.66	3.95
64	s37 99 LC	3.74	0.93	0.93	1.87	13.08	3.74	18.69	14.95	7.48	9.35	11.22	5.61	3.74	0.93	3.74
65	s38 99 LC	1.73	1.73	1.73	3.46	22.51	1.73	24.24	18.18	5.19	4.33	6.93	3.46	2.60	0.43	1.73
66	s39 99 P	3.15	0.79	0.79	1.57	9.45	3.15	17.32	14.17	9.45	7.87	14.17	6.30	6.30	0.79	4.72
67	s40 99 P	3.01	0.75	0.75	1.50	12.03	3.01	19.55	15.04	7.52	6.02	12.03	6.02	6.02	0.75	6.02
68	s41 99 P	4.20	0.70	0.70	2.80	12.59	2.80	16.78	12.59	5.59	6.99	13.99	5.59	8.39	0.70	5.59
69	s42 99 P	2.70	0.68	0.68	1.35	9.46	2.70	16.22	12.16	8.11	6.76	12.16	6.76	14.87	1.35	4.05
70	s43 99 P	2.86	0.71	0.71	1.43	11.43	2.86	18.57	14.29	7.14	7.14	12.86	5.71	7.14	1.43	5.71
71	s44 99 P	2.76	0.69	0.69	1.38	11.03	2.76	19.31	15.17	5.52	6.90	13.79	5.52	8.28	0.69	5.52
72	s45 99 P	2.08	0.52	0.52	4.17	11.46	20.83	12.50	10.42	5.21	5.21	10.42	4.17	6.25	1.04	5.21
73	s46 99 P	4.03	0.67	0.67	2.68	12.08	4.03	18.79	14.77	8.05	8.05	10.74	5.37	4.03	0.67	5.37
74	s49 99 C	2.51	0.42	1.67	2.51	16.74	5.02	14.23	15.06	6.69	8.37	10.04	6.69	5.86	0.84	3.35
75	50A99 C	12.80	0.80	0.80	3.20	12.80	3.20	16.00	14.40	8.00	6.40	9.60	4.80	3.20	0.80	3.20
76	s52 99 C	3.95	0.66	2.63	2.63	15.79	5.26	18.42	14.47	5.26	7.89	9.21	5.26	5.26	0.66	2.63
77	s53 99 C	1.94	0.97	0.97	1.94	14.56	4.85	18.45	15.53	5.83	8.74	10.68	5.83	4.85	0.97	3.88
78	55A99 C	3.54	0.88	0.88	1.77	14.16	3.54	19.47	15.93	5.31	8.85	10.62	5.31	5.31	0.88	3.54
79	s56 99 C	3.42	0.85	0.85	1.71	11.97	5.13	18.80	13.68	6.84	10.26	11.97	5.13	5.13	0.85	3.42
80	s57 99 C	1.75	0.88	0.88	1.75	12.28	3.51	15.79	14.04	8.77	7.02	12.28	5.26	8.77	1.75	5.26
81	s58 99 C	3.42	0.85	0.85	1.71	8.55	3.42	18.80	15.39	8.55	8.55	11.97	5.13	8.55	0.85	3.42

Appendix 5. Concentrations (%) of PAHs in samples with 0.5<SPAH<2 mg/kg

	mean	std	cv	median	max	min
N	2.79	1.87	67.19	2.63	12.80	0.58
Acy	0.66	0.25	38.08	0.68	1.73	0.08
Ace	1.77	1.51	85.72	1.48	8.82	0.52
Fen	2.44	1.02	41.53	2.11	5.49	1.30
Phe	13.35	3.80	28.49	12.24	29.07	4.89
Ant	4.89	5.10	104.31	3.94	44.57	1.36
Flu	18.41	3.76	20.42	18.31	28.57	6.86
Pyr	14.71	2.63	17.90	14.71	25.18	5.71
BaA	6.83	1.41	20.57	6.84	9.89	2.61
Chr	7.89	1.70	21.52	7.88	15.07	2.71
BFl	11.46	2.62	22.85	11.86	18.33	3.29
BaP	5.58	1.32	23.72	5.63	9.21	1.55
ind	4.59	2.10	45.72	4.35	14.86	1.36
DahA	0.95	0.81	85.18	0.82	7.43	0.19
ghi	3.68	1.15	31.39	3.54	6.67	0.78

Appendix 6. Concentrations (%) of PAHs in samples with SPAH > 2.0 mg/kg

	mean	std	cv	median	max	min
N	2.97	2.55	85.82	1.92	10.09	0.58
Acy	0.51	0.31	60.23	0.49	1.26	0.08
Ace	2.14	1.85	86.36	1.41	6.98	0.58
Fen	3.01	1.27	42.22	2.63	4.90	1.74
Phe	15.78	5.53	35.06	15.00	29.07	9.89
Ant	5.09	3.03	59.52	5.00	13.36	1.36
Flu	18.24	3.06	16.79	17.98	23.26	13.81
Pyr	15.16	2.56	16.89	15.69	19.62	11.58
BaA	6.96	1.31	18.76	6.97	9.89	5.04
Chr	7.45	1.85	24.80	7.75	9.28	2.71
BFl	9.77	2.94	30.07	10.56	13.68	3.29
BaP	5.42	1.68	31.04	5.70	7.04	1.55
ind	3.89	1.34	34.58	4.01	6.11	1.36
DahA	0.64	0.29	45.41	0.63	1.26	0.19
ghi	2.96	0.95	32.16	3.21	4.06	0.78

Appendix 7. Correlation coefficients of PAHs, TOC, and fractions (%) of gravel, sand, and silt

	N	Ace	Acy	Fen	Phe	Ant	Flu	Pyr	BaA	Chr	BFl	BaA	ind	Dah	ghi	TOC	gravel	sand	silt	clay
N	1.00	0.51	0.25	0.48	0.40	0.35	0.38	0.34	0.45	0.47	0.45	0.55	0.56	0.35	0.57	0.05	0.38	0.43	-0.45	-0.32
Ace	0.51	1.00	0.41	0.51	0.50	0.26	0.48	0.38	0.60	0.53	0.54	0.49	0.72	0.45	0.64	0.00	0.12	0.21	-0.23	-0.09
Acy	0.25	0.41	1.00	0.88	0.88	0.21	0.61	0.51	0.45	0.32	0.29	0.29	0.29	0.17	0.26	-0.05	-0.03	0.12	-0.04	-0.16
Fen	0.48	0.51	0.88	1.00	0.94	0.54	0.76	0.68	0.68	0.59	0.55	0.57	0.55	0.37	0.55	0.00	0.11	0.23	-0.18	-0.23
Phe	0.40	0.50	0.88	0.94	1.00	0.47	0.77	0.67	0.64	0.54	0.50	0.52	0.54	0.35	0.52	-0.01	0.04	0.19	-0.13	-0.17
Ant	0.35	0.26	0.21	0.54	0.47	1.00	0.55	0.54	0.60	0.62	0.58	0.61	0.59	0.41	0.62	0.03	0.21	0.23	-0.24	-0.19
Flu	0.38	0.48	0.61	0.76	0.77	0.55	1.00	0.98	0.94	0.91	0.89	0.87	0.72	0.60	0.80	0.06	0.00	0.06	0.00	-0.15
Pyr	0.34	0.38	0.51	0.68	0.67	0.54	0.98	1.00	0.93	0.92	0.91	0.90	0.68	0.60	0.79	0.08	0.00	0.04	0.01	-0.14
BaA	0.45	0.60	0.45	0.68	0.64	0.60	0.94	0.93	1.00	0.98	0.97	0.95	0.87	0.69	0.93	0.06	0.03	0.11	-0.08	-0.12
Chr	0.47	0.53	0.32	0.59	0.54	0.62	0.91	0.92	0.98	1.00	0.99	0.98	0.85	0.70	0.94	0.11	0.05	0.14	-0.09	-0.16
BFl	0.45	0.54	0.29	0.55	0.50	0.58	0.89	0.91	0.97	0.99	1.00	0.98	0.85	0.71	0.94	0.12	0.06	0.11	-0.09	-0.13
BaA	0.55	0.49	0.29	0.57	0.52	0.61	0.87	0.90	0.95	0.98	0.98	1.00	0.83	0.70	0.94	0.16	0.11	0.20	-0.17	-0.19
ind	0.56	0.72	0.29	0.55	0.54	0.59	0.72	0.68	0.87	0.85	0.85	0.83	1.00	0.64	0.93	0.02	0.14	0.21	-0.24	-0.06
Dah	0.35	0.45	0.17	0.37	0.35	0.41	0.60	0.60	0.69	0.70	0.71	0.70	0.64	1.00	0.68	0.05	0.07	0.20	-0.14	-0.20
ghi	0.57	0.64	0.26	0.55	0.52	0.62	0.80	0.79	0.93	0.94	0.94	0.94	0.93	0.68	1.00	0.08	0.15	0.20	-0.22	-0.12
TOC	0.05	0.00	-0.05	0.00	-0.01	0.03	0.06	0.08	0.06	0.11	0.12	0.16	0.02	0.05	0.08	1.00	0.33	0.32	-0.32	-0.32
gravel	0.38	0.12	-0.03	0.11	0.04	0.21	0.00	0.00	0.03	0.05	0.06	0.11	0.14	0.07	0.15	0.33	1.00	0.48	-0.70	-0.45
sand	0.43	0.21	0.12	0.23	0.19	0.23	0.06	0.04	0.11	0.14	0.11	0.20	0.21	0.20	0.20	0.32	0.48	1.00	-0.90	-0.68
silt	-0.45	-0.23	-0.04	-0.18	-0.13	-0.24	0.00	0.01	-0.08	-0.09	-0.09	-0.17	-0.24	-0.14	-0.22	-0.32	-0.70	-0.90	1.00	0.45
clay	-0.32	-0.09	-0.16	-0.23	-0.17	-0.19	-0.15	-0.14	-0.12	-0.16	-0.13	-0.19	-0.06	-0.20	-0.12	-0.32	-0.45	-0.68	0.45	1.00

Appendix 8. PAH concentrations (mg/kg dry weight) in the standard reference sediments HS5 and EC2 and their 90% confidence limits (CL)

PAH	HS5	HS5+CL	HS5-CL	EC2	EC2CL	EC2-CL	PAH
N	0.25	0.32	0.18	1.47	2.57	0.37	N
Acy	0.15	0.15	0.15	0.12	0.17	0.07	Acy
Ace	0.23	0.33	0.13	0.2	0.28	0.12	Ace
Fen	0.4	0.5	0.3	2.14	2.94	1.34	Fen
Phe	5.2	6.2	4.2	1.41	1.72	1.1	Phe
Ant	0.38	0.53	0.23	0.11	0.16	0.06	Ant
Flu	8.4	11	5.8	3.55	4.38	2.72	Flu
Pyr	5.8	7.6	4	2.92	3.55	2.29	Pyr
BaA	2.9	4.1	1.7	1.42	1.93	0.91	BaA
Chr	2.8	3.7	1.9	3.6	5.07	2.13	Chr
BaP	1.7	2.5	0.9	4.46	6.04	2.88	B(b+k)Fl
B(b+k)Fl	3	4.4	1.6	1.91	2.63	1.19	BeP
ghi	1.3	1.6	1	1.21	1.77	0.65	BaP
DahA	0.2	0.3	0.1	1.55	2.08	1.02	ind
ind	1.3	2	0.6	1.47	2.12	0.82	ghi
				0.49	0.7	0.28	DahA
sum	34.01	45.167	22.853	28.03	38.11	17.95	SPAH
Horwitz	68			65			

Appendix 9. Deviations in the analyses of standard reference sediments, certified minus found concentrations in percent of certified concentrations

	1996	1998	1999
N	32	-159	-30
Acy	87	42	6
Ace	83	85	-30
Fen	63	57	-29
Phe	37	1	-20
Ant	18	-155	20
Flu	18	6	-1
Pyr	34	9	-19
BaA	51	32	-21
Chr	3	49	19
B(b+k)Fl	0	-51	30
BaP	49	32	-10
ind	24	-5	30
DahA	30	43	30
ghi	40	4	-19

Appendix 10. PAHs (mg/kg) in additional samples from Rodney Marginal, collected in June 1999 in the vicinity of stations identified in Appendices 1-4 as 3, and 57-60, and three other stations not considered previously because of their SPAH < 0.5. Non-detectable levels are replaced by one half of the detection limit (0.005 mg/kg)

Append 1 #	3				Addit.	Addit.	Addit.	Addit.	57	58	59	60
PAH	3s16rm	96sjpc-17	98sjpc-14	99sj23	sa#59	sa#60	sa#61	sa#62	s19 99 RM	s20 99 RM	s21 99 RM	s25 99 RM
N	0.01	0.02	0.01	0.005	0.03	0.005	0.03	0.005	0.03	0.04	0.01	0.04
Acy	0.005	0.005	0.005	0.005	0.01	0.005	0.005	0.005	0.02	0.005	0.005	0.01
Ace	0.06	0.01	0.005	0.005	0.005	0.005	0.005	0.005	0.36	0.02	0.005	0.03
Fen	0.06	0.01	0.01	0.005	0.02	0.005	0.02	0.005	0.25	0.11	0.01	0.03
Phe	0.26	0.05	0.03	0.04	0.18	0.02	0.11	0.06	1.5	0.47	0.07	0.19
Ant	0.04	0.01	0.01	0.005	0.05	0.005	0.02	0.005	0.07	0.3	0.04	0.05
Flu	0.24	0.07	0.04	0.05	0.28	0.04	0.11	0.06	1.2	0.31	0.06	0.25
Pyr	0.19	0.05	0.04	0.04	0.23	0.03	0.08	0.05	0.96	0.26	0.05	0.22
BaA	0.06	0.02	0.01	0.03	0.11	0.005	0.05	0.02	0.26	0.16	0.07	0.08
Chr	0.07	0.03	0.02	0.02	0.11	0.005	0.05	0.005	0.14	0.15	0.11	0.09
BFl	0.08	0.05	0.03	0.04	0.16	0.03	0.11	0.05	0.17	0.16	0.12	0.1
BaP	0.04	0.03	0.01	0.02	0.07	0.005	0.04	0.005	0.08	0.09	0.06	0.04
ind	0.03	0.02	0.02	0.03	0.07	0.005	0.1	0.005	0.07	0.09	0.07	0.04
daha	0.005	0.005	0.005	0.005	0.005	0.005	0.005	0.005	0.01	0.01	0.01	0.005
ghi	0.02	0.02	0.01	0.02	0.05	0.005	0.02	0.02	0.04	0.07	0.04	0.02
SPAH	1.17	0.40	0.26	0.32	1.38	0.18	0.76	0.31	5.16	2.25	0.73	1.20
SPAH estd	1.27	0.49	0.35	0.40	1.45	0.35	0.67	0.44	5.69	1.59	0.44	1.32
Horw	103	113	120	119	101	130	107	124	88	93	107	100

Appendix 11. Dredging sample grid at Rodney Marginal. Blocks are 22.5 by 22.5 m

9 Sample 59 SPAH 0.73	8 Sample 17 SPAH 0.40 Sample 58 SPAH 2.25	7 Sample 14 SPAH 0.26	6	5	4 X Additional Samples SPAH 1.38, 0.18, 0.76, 0.31	3 Sample 57 SPAH 5.16	2 Sample 3 SPAH 1.17
37	36	35 Sample 60 SPAH 1.20	34	33	32	31	30 Sample 99sj23 SPAH 0.32