

Spatial Distribution of Polychlorinated Biphenyls in Coastal Marine Sediments Receiving Industrial Effluents in Kuwait

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Abstract. The concentrations of polychlorinated biphenyls (PCBs) were measured in surficial sediments receiving industrial and municipal effluents in Kuwait. The Σ PCB concentrations varied by two orders of magnitude ranging from 0.4 to 84 $\mu\text{g kg}^{-1}$ dw. The homologue distribution in the study favored the more chlorinated congeners and generally followed the order: penta-PCBs > hexa-PCBs > tetra-PCBs \approx hepta-PCBs, with the dominant congeners being 138, 101, 110, 180, 153, 132, 149, and 118. The spatial distribution revealed significant intersite difference in concentration, with high levels encountered close to a harbor and several wastewater outlets suggesting that point source input is the primary delivery mechanism of PCBs to the sediment. This study suggests that atmospheric deposition of PCBs may not be a significant delivery mechanism to sediments in Kuwait possibly due to low annual precipitation and high annual temperatures that are experienced in the Arabian Gulf. The implication of this observation is that PCBs in air are likely to remain in the gas phase long enough to be subject to long-range atmospheric transport to other regions.

The discovery of oil in the Arabian Gulf, and in Kuwait in particular, led to rapid urbanization, population growth, industrialization and with it, several pollution problems from oil and non-oil sources. Major industries in the Gulf States are located along the coast. These include the heavy non-oil industries such as power and desalination plants, petrochemical plants, refineries, etc. This has led to the introduction of a variety of chemicals including heavy metals, oil and petroleum-related compounds, nutrients, and halogenated organics into coastal marine sediments.

Polychlorinated biphenyls (PCBs) are a class of industrial chemicals that were used extensively in electrical transformers and capacitors as heat transfer fluids, and in consumer products (Edujje 1988; Harrad *et al.* 1994). They are ubiquitous in the environment and can enter the marine environment from a variety of sources including (i) direct deposition from the atmosphere, (ii) runoff from land, and (iii) directly from

industrial and wastewater treatment plant discharges (Broman *et al.* 1996; Preston 1992; Thompson *et al.* 1996). They are hydrophobic and, upon entry into the aquatic environment, they rapidly associate with suspended particulate material and sediments. Their ultimate fate depends on a variety of physical, chemical, and biological interactions including biotic uptake, abiotic degradation, volatilization, and burial in bottom sediments (Calamari 2002; Hong *et al.* 1999). PCBs are of environmental concern because they resist degradation by physical, chemical, or biological pathways, bioaccumulate through the food web (Kucklick *et al.* 1996; Muir *et al.* 1988; Thomann 1989), and pose a risk causing adverse effects on human health and the environment (Focant *et al.* 2003; Jones and de Voogt 1999; McDonald 2003).

Most of the studies that have been carried out in Kuwait, to date, have focused on oil and oil-based pollutants (Al-Sulaimi *et al.* 1993; Literathy *et al.* 1992; Massoud *et al.* 1996; Readman *et al.* 1992; Saeed *et al.* 1998), particularly the distribution of polycyclic aromatic hydrocarbons and metals (Al-Yakoob and Saeed 1994; Massoud *et al.* 1998; Saeed *et al.* 1995), in marine sediments. Studies on several classes of persistent organic pollutants have up until now received very little attention. This was, in part, a result of lack of appropriate analytical facilities and expertise in most Gulf States (Fowler 2002). Some attempts had been made to redress this balance, by the inclusion of some of these chemicals in national monitoring efforts. These data are, however, not accessible to the wider scientific community. This study reports the concentrations and spatial distribution of PCBs in coastal sediments facing an industrial estate in Kuwait, and examines the potential input mechanisms of these compounds to the sediment.

Materials and Methods

Study Area

The Shuaiba Industrial Area (SIA) is located approximately 50 km south of Kuwait City and faces a 12.5-km stretch of sea. The total area of the estate is about 23 million square meters and is divided into the eastern and western sectors (Figure 1). There are over 35 different industries in the mainly eastern sector comprising three refineries, two power generation plants, an industrial gas corporation, a major harbor,

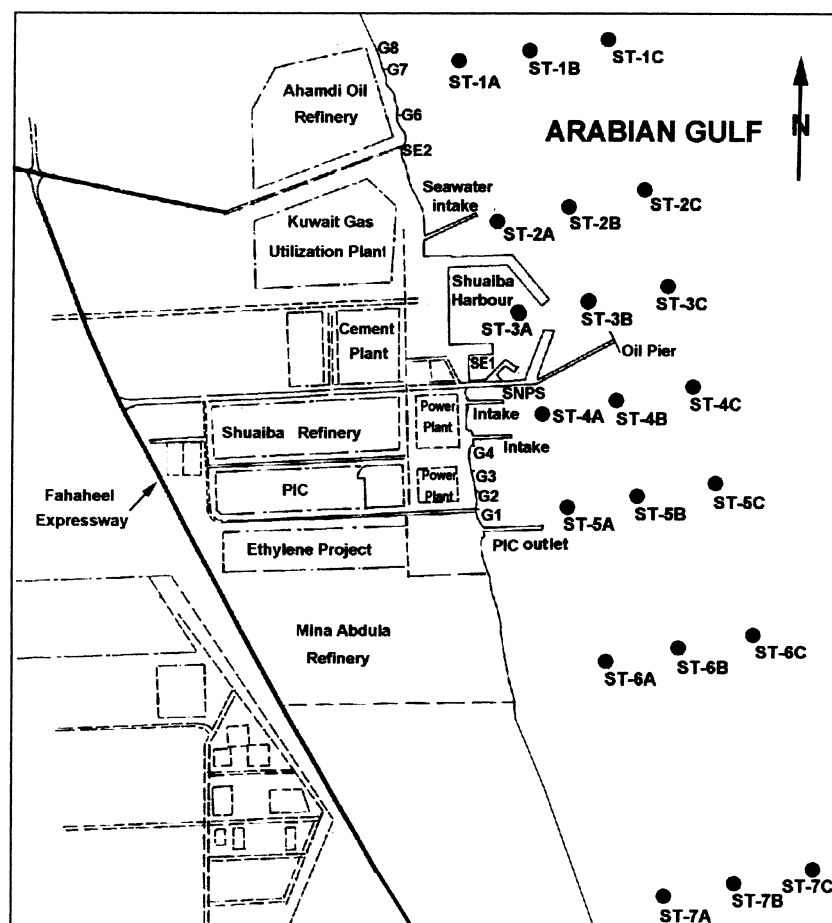


Fig. 1. Map showing sampling locations along the coastline facing the industrial estate.

a cement factory, two petrochemical companies, a paper production company, and a polypropylene plant, to name a few. These generate in excess of $31,000 \text{ m}^3 \text{ day}^{-1}$ of wastewater, of which roughly 95% is collected and discharged to the sea. In addition to sea cooling water returns, there are two main types of wastewater generated at the estate: industrial wastewater and sanitary wastewater. Industrial wastewater from process sources and contaminated surface drainage is collected and discharged to the sea either directly or via the common storm water drainage system. Sanitary wastewater is collected in septic tanks, treated in activated sludge plants, and chlorinated before discharge to the sea.

Sample Collection

Surficial sediment samples were collected from 22 stations (Figure 1) along a 12.5-km stretch of coastline facing the SIA using a van Veen grab sampler. Table 1 gives the location and depth information of the sampling stations. The near-shore water of the SIA is shallow, with an average depth of 5 m up to 1 km offshore. As a consequence, hydrophobic pollutants discharged to the sea might not be well mixed and might have a short water column residence time. The strategy was to collect samples along seven transects, six of which face a major industry and are close to a wastewater discharge point, at 0.5, 1 and 1.5 km from the shoreline. Transect 7 was chosen as a background site because it is further removed from any known discharge points. A single sample was collected further south, away from the influence of the major industrial site. At each site, three samples were collected in a triangular array within a 5 m radius, the top 2 cm pooled, before

subsamples were obtained for chemical analyses. The samples were immediately transferred into clean, solvent rinsed, amber glass jar and stored in a cool box for transport to the laboratory where they were kept at -25°C prior to analysis. All utensils used in the collection, pooling, and subsampling were thoroughly washed and rinsed with acetone and hexane between sampling sites to minimize the likelihood of cross-contamination.

Chemicals and Reagents

All solvents used in this work were analytical grade and purchased from Fisher Scientific. Silica (100–200 mesh), alumina, and sodium sulfate were purchased from VWR Scientific. The PCB analytical standard (EC-4133) was purchased from Cambridge Isotope Laboratory (CIL, Andover, MA). The following IUPAC congeners were in the standard mix in their elution order off a DB 5 ms column: 18, 17, 31, 28, 33, 52, 49, 44, 74, 70, 101, 99, 87, 110, 151, 149, 118, 153, 105, 138, 132, 187, 183, 128, 177, 171, 156, 180, 191, 169, 170, 201, 208/195, 194, 205, 206, and 209. Mirex, used as the internal standard, was purchased from CIL. The recovery standard, EC 4058, containing $^{13}\text{C}_{12}$ -PCBs 28, 52, 101, 153, 138, 180, and 209, was purchased from CIL.

Sediment Characterization

The sediment samples were characterized for particle size distribution, organic matter content, and percent organic carbon (OC). Detailed experimental procedures are presented elsewhere (Beg *et al.* 2001).

Table 1. Information on sampling location and water depth

Sample code	Major activity	Distance from shore (km)	Latitude (°N)	Longitude (°E)	Depth (m)
1A	Refinery (Mina Al-Ahmadi)	0.5	29 03.763	048 09.411	7
1B		1	29 03.758	048 09.918	18
1C		1.5	29 03.770	048 10.222	21
2A	Gas utilization plant	0.5	29 03.174	048 09.271	5
2B		1	29 03.086	048 09.638	12
2C		1.5	29 02.811	048 10.093	19
3A	Shuaiba harbor	0.5	29 03.076	048 09.643	9
3B		1	29 02.381	048 09.579	16
3C		1.5	29 02.797	048 10.106	17
4A	Power plant	0.5	29 02.214	048 09.649	3
4B		1	29 02.213	048 10.305	8
4C		1.5	29 02.367	048 10.847	12
5A	Petrochemical industry	0.5	29 01.895	048 09.912	4.5
5B		1	29 01.924	048 10.380	7
5C		1.5	29 01.902	048 10.732	12
6A	Refinery (Mina Abdullah)	0.5	29 01.362	048 10.413	4
6B		1	29 01.334	048 11.117	5
6C		1.5	29 01.338	048 11.149	9
7A	Boundary transect	0.5	29 00.149	048 09.925	3
7B		1	29 04.280	048 09.322	5
7C		1.5	29 00.144	048 09.905	5

Extraction and Analyses

Sediments for PCB analyses (15–20 g) were mixed with anhydrous sodium sulfate that had been previously baked out at 450°C to remove residual water. The mixture was placed in pre-extracted thimbles and Soxhlet extracted for 12–16 hours using hexane. Prior to extraction, the samples were spiked with a recovery standard containing $^{13}\text{C}_{12}$ -PCBs 28, 52, 101, 153, 138, 180, and 209. Copper turnings were added to the flasks during the extraction stage to remove elemental sulphur. The extracts were concentrated to about 0.5 ml using a Turbovap evaporator (Zymark Corp., Hopkington, MA) and chromatographed on a silicic acid/alumina column (10 g/5 g, respectively, in a 19 mm i.d. column) to remove polar interferences. The PCBs were eluted with 100 ml hexane:dichloromethane (1:1 (v/v)) after discarding the initial 20 ml eluting off the column. The eluent was reduced in volume to 0.5 ml in a Turbovap evaporator and further cleaned on a liquid–solid chromatographic column (9 mm i.d.) containing 2 g of silica gel (Merck, 60–230 mesh), 1 g of alumina (BDH neutral alumina), all baked at 450°C overnight. These were slurry packed in hexane and capped with approximately 0.2 g of anhydrous sodium sulfate to prevent the column from going dry or trapping air bubbles. The silica gel used throughout this work was pre-extracted with hexane for 24 hours, changing solvents every 6 hours, dried under vacuum at 90 mbars of pressure, and stored in solvent-rinsed glass jars lined with solvent-rinsed aluminum foil. The compounds were eluted with 40 ml hexane:DCM (1:1 v/v) blown down under a gentle stream of nitrogen. Fifty microliters of dodecane were added during the blow-down stage to ensure that the samples did not dry out and transferred to 100- μl glass inserts, and internal standards (mirex and PCB 204) were added just prior to GC analysis.

Instrumental Analysis

The sample extracts were analyzed with an Agilent 6890N gas chromatograph using splitless injection on a 30 m HP5–ms column (0.25 mm i.d., 0.25 μm film thickness) and helium as carrier gas. The oven program was 150°C for 2 min, ramped at 30°C/min to 170°C, 4°C/min to 200°C and held for 13 min, and further ramped at 3°C/min

to 280°C and held for 10 min. This was coupled to an Agilent 5973 inert mass selective detector, operated in NCI mode (using selected ion monitoring), with methane as reagent gas. Operating conditions were as follows: injector temperature was set at 265°C; ion source 230°C; quadrupole 106°C; transfer line 290°C. Identification and quantification was carried out against five calibration standards of known concentration. A peak was positively identified if it was within ± 0.05 min of the retention time in the calibration standard and quantified only if the S/N = 3, and the ratio of the ion to its qualifier ion was within $\pm 20\%$ of the standard value. The concentrations of PCBs were calculated by dividing the masses by the dry weight (dw) of sediment, determined gravimetrically after drying separate sub-samples of the sediment to constant weight at 105°C. ΣPCBs is defined as the sum of 26 congeners (74, 77, 101, 99, 87, 110, 151, 149, 118, 153, 105, 138, 132, 187, 183, 128, 177, 171, 156, 180, 170, 201, 195, 208, 194, 206, and 209).

QA/QC

An analytical blank was processed for every five samples with the same weight of sodium sulfate used to dry the sediments. The PCBs present in the appropriate blank were subtracted from those in the sample extracts. Sample peaks are reported only if the signal exceeded three times the baseline noise. Procedural blanks were used to determine the detection limits (LOD = mean blank + 3 \times SD). The detection limits are in the range of 10 pg/g. Average recoveries (%) \pm 1 standard deviation of ^{13}C -surrogates were as follows: ^{13}C -101, 78 \pm 15%; ^{13}C -153, 82 \pm 18%, ^{13}C -138, 75 \pm 10%; ^{13}C -180, 106 \pm 11%; ^{13}C -101, 98 \pm 8%. The accuracy and precision of the method employed in the analysis were assessed by extracting three marine sediment reference materials that have been certified for certain PCB congeners. The results obtained were within 6–15% of the quoted value for >95% of the certified congeners in the three reference materials.

Results and Discussion

The concentration data for PCBs in sediments arising from this study are presented in Table 2. The ΣPCB concentration in the

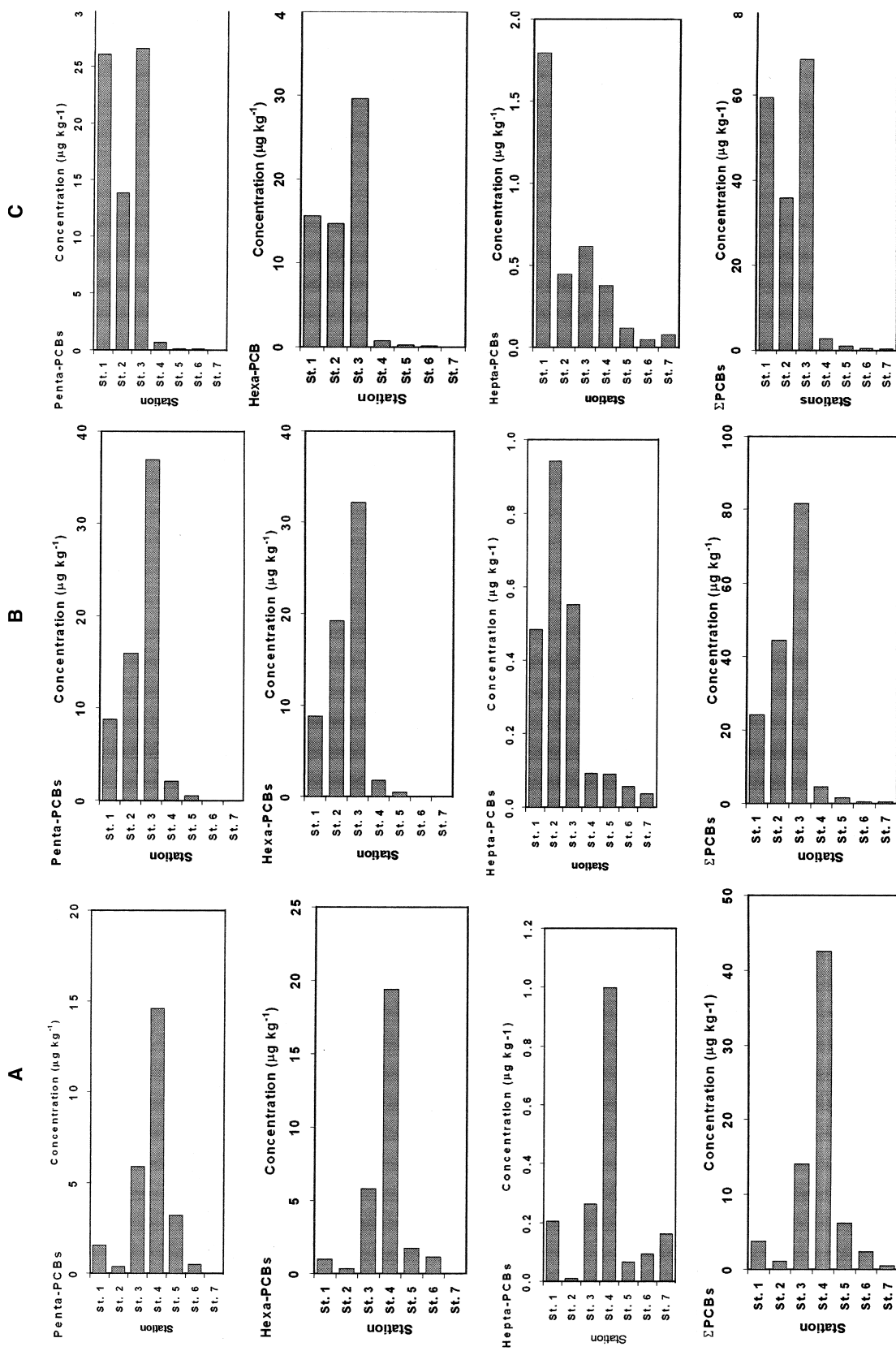


Fig. 2. Concentrations of homologs and ΣPCBs (µg kg⁻¹ dw) in sediments. (A–C) Sampling stations located at 0.5, 1.0, and 1.5 km from the shoreline, respectively. The stations 1–7 are arranged in a north–south order within the study area, with station 1 being the farthest north and station 7 the farthest south.

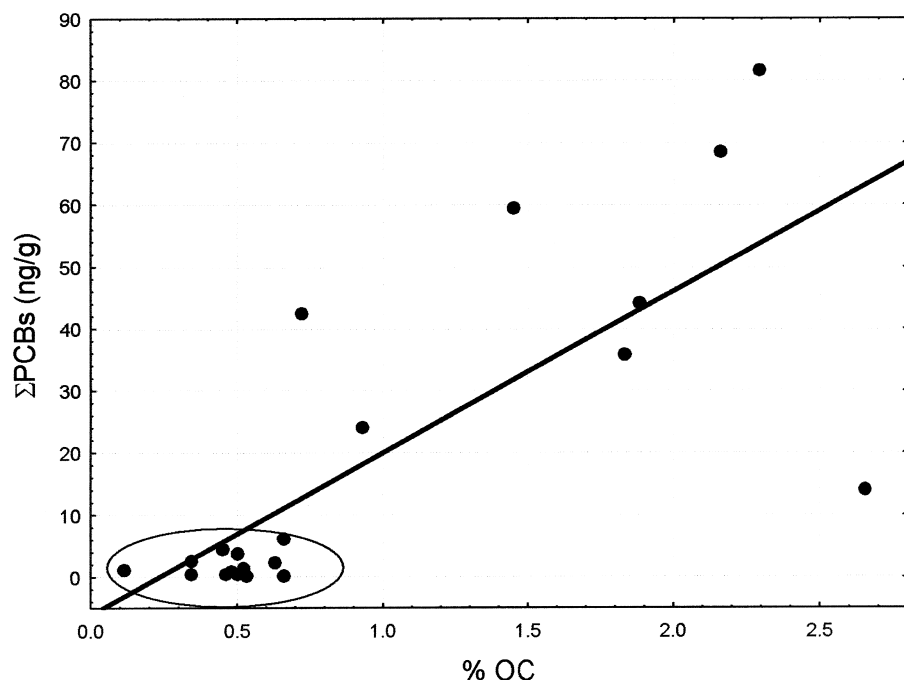


Fig. 3. Scatter plot of Σ PCBs ($\mu\text{g kg}^{-1}$) and the percent organic carbon of sediments.

The nearshore water in the study area is very shallow, averaging about 7 m in depth up to 1 km away from the shoreline. This will suggest that pollutants entering the sea from land-based sources, *e.g.*, wastewater or surface runoff, will very quickly settle to the bottom, resulting in higher concentrations nearshore and an exponential decrease seaward. The distribution pattern observed in this study suggests that the distribution of PCBs are controlled by other overriding factors such as wave action, organic carbon content of the sediments, and, in part, sediment focusing (Swackhamer and Eisenreich 1991; Wong *et al.* 1995), a process whereby fine sediments are transported by current from nearshore shallower portions to deeper waters carrying with it its pollutant load.

Organic Carbon

The percent OC of the samples varied between 0.11 and 2.65% with an arithmetic mean of $0.96 \pm 0.021\%$ Beg *et al.* 2001). The highest OC values were recorded at station 3A, in front of the harbor used for loading oil onto ships. The average OC values recorded are typical of values reported for the entire Gulf (Massoud *et al.* 1996). The high OC values may be related, in part, to releases of oil during loading operations and wastewater from the oil refinery process.

The spatial distribution of PCBs tracks the OC content of sediments closely, although only a weak linear relationship exists between the two parameters. Figure 3 is a scatter plot of Σ PCBs vs. percent OC. It is clearly evident that the stations with low OC content correspondingly had low Σ PCB concentrations, and high PCB concentrations were encountered at stations with moderate to high OC content. The concentration of hydrophobic contaminants depends to a large extent on the retaining capacity of the sediment because their sorptive properties are largely controlled by their hydrophobicity and by the particle OC content (Schwarzenbach *et al.* 1991). The

OC content of the sediments at some sites was extremely low, thereby limiting their ability to retain the compounds.

When principal component analysis was applied to the data, two distinct groups could be made out (Figure 4). Group 1 included stations with high PCB concentrations and high OC content, whereas those stations with relatively lower PCB concentrations and OC content tended to cluster together. Furthermore, the congener distribution suggests that the penta- and hepta-PCB congeners dominated the mixture, suggesting that the low-molecular-weight, more aqueous soluble congeners are either not reaching the sediment or are poorly preserved.

Variation of Σ PCB Concentration with Depth of Sampling

The concentrations of PCBs measured at the various sites appear to be linked to the depth of sampling as can be seen in the scatter plot in Figure 5. The plot suggests that, by and large, the shallower the sampling station, the lower the Σ PCB concentration and *vice versa*. Incidentally, also, the grain size distribution suggests that the deeper portions are slightly dominated by sediments in the 63- μm size range relative to the distribution in the shallower portions where coarser material is dominant. This would appear to suggest that the deeper areas in the study area are depositional zones where the likelihood of sediment focusing is high.

The spatial distribution observed in the study area cannot be explained by a single factor. Rather, a combination of factors including: point source input, OC content, and post-depositional mobility by wave action leading to focusing of sediments with their associated pollutant load is controlling the observed "spotty" distribution. The correlation coefficients in Table 4 seem to support the notion that pollutants are associated with fine-grained sediments with relatively high OC content. The low PCB concentrations southward from

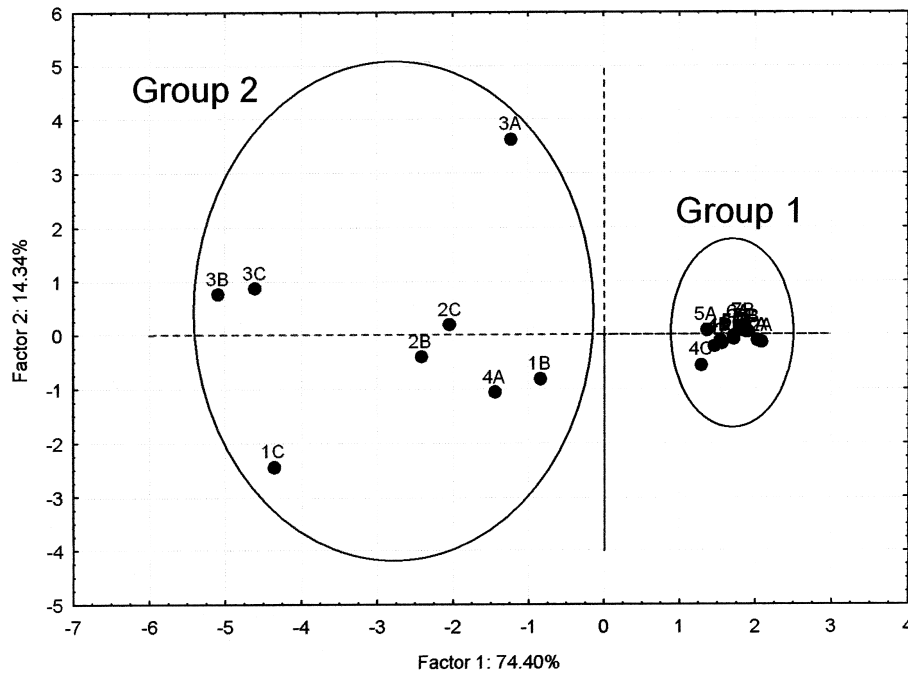


Fig. 4. PCA score plots for samples from the study area for Σ PCBs, organic carbon, and depth of sampling. Principal components 1 and 2 accounted for from 74.4% and 14.34%, respectively.

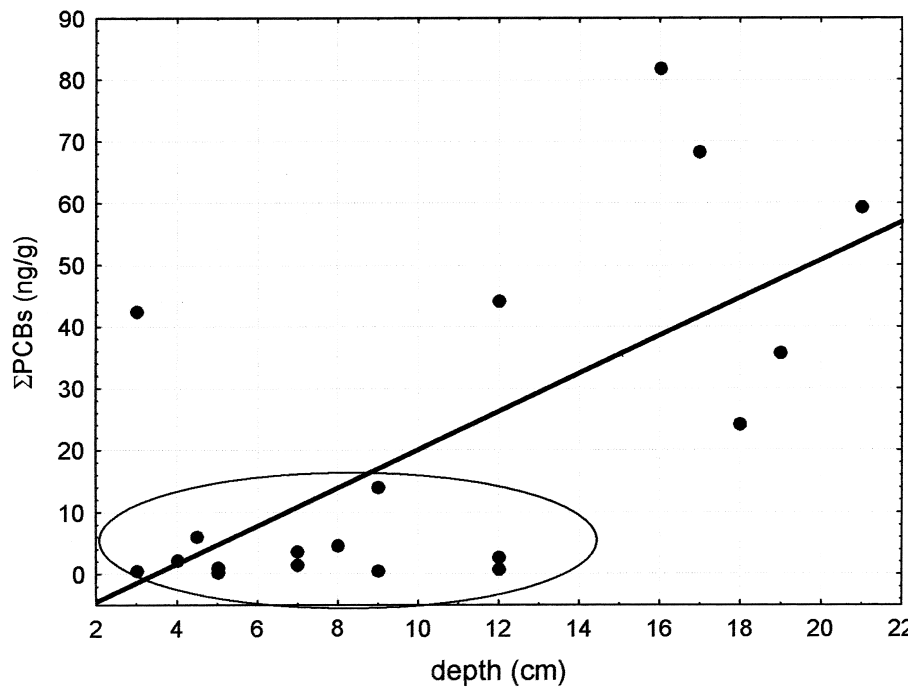


Fig. 5. Scatter plot of the variation of Σ PCB content of sediment and the water depth at the sampling site.

station 4 may be because the wastewater discharged at these points may not be from industries using and/or forming the compounds in their operations and as such these levels may represent background levels. A shoreline survey of sediments from Kuwait's coastline carried out after the 1991 Gulf War also revealed that localized "hot spots" of PCBs exist in the vicinity of a desalination plant and a shipping port (IAEA 1998). This supports the notion that land-based sources are the most important delivery vehicles for PCBs to sediments in the region.

Implications for Global Distribution of PCBs

It is obvious from the observed distribution that land-based sources are the major delivery mechanism of PCBs to coastal sediments in Kuwait and, based on evidence from other studies, possibly sediment from the entire Arabian Gulf. Atmospheric deposition, which is known to play a very significant role in delivering PCBs to water bodies in Europe and North America, is not significant in this region. This observation may be due to the extremely high temperatures expe-

Table 4. Correlation matrix for PCB concentrations with depth, % OC, and % grain size (< 63 µm)

	% Organic carbon	Sampling depth (m)	% Grain size (< 63 µm)	ΣPCBs (µg kg ⁻¹)
% Organic carbon	1			
Sampling depth (m)	0.60	1		
% Grain size (< 63 µm)	0.86	0.31	1	
ΣPCBs (µg kg ⁻¹)	0.75	0.67	0.49	1

rienced for the greater part of the year in the Arabian Gulf coupled with extremely low annual precipitation. Both of these factors do not support deposition of PCBs, 95% of which is in the vapor phase at 25°C (Falconer *et al.* 1995; Hornbuckle and Eisenreich 1996; Manchester-Neesvig and Andren 1989), well below the annual average temperatures experienced in the Middle East.

In Kuwait, for example, average annual temperatures are about 35°C (range 10–50°C) and average annual rainfall ranges between 78 and 152 mm, occurring between November and February. The vapor pressure of PCBs (Duinker and Bouchertall 1989; Pankow and Bidleman 1991, 1992) suggests that they almost entirely exist in the gas phase at the temperatures experienced in the region for the better part of the year. Furthermore, because atmospheric removal, via wet and dry deposition, is more efficient for particulate-bound compounds relative to vapor phase compounds (Arimoto 1989; Brorstrom-Lunden 1996; ter Schure *et al.* 2004), any form of deposition in this region will favor high-molecular-weight compounds. Vapor exchange across the air–water interface will probably be insignificant. The low-molecular-weight ones, predominantly in the gas phase, will have a higher atmospheric residence time and will be subject to other processes, like long-range atmospheric transport to other regions and/or gas phase reactions (Wania and Mackay 1996), like OH radical removal or photolysis.

The significant intersite difference in concentration in the study area is further evidence implicating point source input as the primary delivery mechanism of PCBs to the sediment. The most likely source is wastewater discharged from the various discharge points along the shoreline. In conclusion, the observed distribution pattern suggests that the principal delivery mechanism for PCB to coastal sediments in this study is from point source inputs.

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