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Fate and transport of planar and mono-*ortho* polychlorinated biphenyls and polychlorinated naphthalenes in Southern California sediments

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Introduction

Polychlorinated biphenyls (PCBs) are an important class of industrial pollutants that are ubiquitous in the marine and freshwater environments (NAS, 1979). Although the “planar” or “non-*ortho*” PCB congeners are present in much lower concentrations than other PCB congeners, they can be responsible for much of the dioxin-like (TCDD) toxicity in the marine ecosystem. Further, their environmental fate may be different from other PCBs. Their determination is, therefore, crucial for assessment of contaminant status of environmental compartments. Mono-*ortho* PCBs and polychlorinated naphthalenes (PCN) although less potent, also exhibit TCDD-like toxicity. Yet, only total PCBs are usually reported and the levels of non- and mono-*ortho* PCBs relative to total PCBs have rarely been studied and data on PCN are uncommon in oceanic samples (Jarnberg et al., 1993). Among the known sources, municipal waste dischargers could be a significant contributor of these synthetic compounds to the ocean (i.e., Venkatesan, 1998; Zeng and Venkatesan, 1999).

The pollutants loads discharged into the Santa Monica (SM) and San Pedro (SP) Basins have changed after the termination of the Hyperion 5-mile outflow for solid waste in late 1987 and from other periodic improvements in the waste water treatment practice. Historic profiles of selected contaminants in the region have been documented using dated sediment cores because sediments integrate and retain records of toxic pollutants influx. Further, sediment samples are well-suited for studies of patterns of relatively stable chemicals, since they reflect the original composition of these substances to a greater extent than biological samples. Such studies of sedimentary chemistry have resulted in stringent laws as well as improved technologies in curtailing waste disposal into the ocean waters, in particular, of toxic chemical contaminants. One of the objectives of this project was to document these changes in the region by measuring the temporal and spatial distribution trends of selected toxic contaminants (i.e., PCBs and polychlorinated naphthalenes) in sediment cores and surface sediments from SM and SP basins and from the SM Bay. This will help understand the fate and transport of these sedimentary contaminants.

The “planar” or “non-*ortho*” PCB congeners (i.e., BZ 77) are strong arylhydrocarbon hydroxylase inducers and endocrine disruptors. Based on their toxicity, they can be considered as one of the real threats to the wildlife and human alike. Studies indicate that the levels of some individual planar PCBs expressed as 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) toxic equivalents equal or precede that of polychlorinated dibenzo-p-dioxins (Safe, 1990). A few recent studies have, therefore, focused on the ‘planar’ PCBs in different environmental compartments such as sediments, fish tissues etc. (i.e., Tanabe et al., 1987; Hong and Bush, 1990; Sericano et al., 1991; Gardinali et al., 1996). They have used carbon column to separate PCBs according to the number of *ortho*-chlorines. By combining with a sulphuric acid cleanup to

remove matrix interference, separation and identification of individual planar PCB congeners at the level of pg/g of sediment has been achieved. Measurement of planar PCBs should provide a more realistic toxicity of the environment rather than a general suite of selected PCBs traditionally measured by many of the environmental monitoring programs. Mono-*ortho* PCBs (i.e., BZ 105), although less potent, also exhibit TCDD-like toxicity, but at a reduced level than planar PCBs (Kannan et al., 1993). We therefore, attempted in the current study to quantitate the coplanar, mono-*ortho* and other PCBs including di-*ortho*PCBs.

Our first and immediate goal was to measure the absolute amounts and the relative ratios of the most toxic “coplanar” to the total PCBs in order to construct their historic profiles in the sediment cores. The second objective was to assess the spatial distribution of these PCBs in the Santa Monica Bay sediments. The overall long term objective was to integrate these current data with our prior findings on other contaminants from the same suite of samples and synthesize into a report on the contaminant status of the region useful for environmental regulatory and policy making decisions.

Sampling locations

Sections from five of the six cores of known Pb-210 chronology from the SM and SP Basins which were analyzed for total PCBs as part of the prior NOAA study (Huh,1998; Venkatesan, 1998) were reanalyzed for PCB speciation from their archived extracts (Figure 1). Six to eleven sections from each of the cores were chosen based on their Pb-210 date to cover from the early 1900s to ~1991. The availability of sediment horizons of 0.5 cm cuts corresponding to 2 to 4 years of sedimentation and their undisturbed surface sediment recovery provided a unique opportunity to follow the historic PCBs distribution in detail in the cores. About 40 out of 75 surface grab sediment samples collected from the Santa Monica Bay in 2000, 2001 and 2002 by the Hyperion personnel were also analyzed (Figure 2).

Methodology

We adapted published methodologies (Sericano et al., 1991; Gardinali et al., 19960, fine-tuned and optimized the protocol to isolate and quantitate the target compounds in the sediment samples using column chromatography with activated carbon and collected discrete cuts of “planar” mono-*ortho* and di-*ortho* PCBs using a mixture of standard PCBs. Then this method was applied to the aromatic fractions archived from prior projects as follows: The total PCBs fractions from several sections from the cores were treated with conc. sulfuric acid to remove matrix interference and subjected to the above column chromatography. Individual PCBs in the three different cuts collected from the column were quantitated using a gas chromatograph equipped with dual column and electron capture detectors and compared with data obtained from previous projects for the same sediment samples (which were processed without the sulphuric acid treatment, Cf: Venkatesan, 1998). The aromatic fractions from the surface sediment samples collected from the Santa Monica Bay were also processed and analyzed similarly to investigate the spatial distribution of the PCB congeners. Laboratory procedure blanks, matrix spike and some duplicate analyses were conducted with every batch of samples as QA/QC measures.

Modifications from the original proposal:

The optimization of the carbon column method took much longer than expected, especially because the specific carbon used in the published procedures is no longer manufactured and we had to try different brands before achieving desired separations.

Concentration of di-*ortho* PCBs was nearly the same in some samples whereas those of mono-*ortho* and planar PCBs were less by ~25% and 75% respectively in all the field samples after sulfuric acid treatment compared to the values obtained in the same sediment samples analyzed in the earlier projects without the acid treatment. However, the recovery of standards was not affected by the acid treatment in the matrix spikes. The much lower levels of planar PCBs in the current analysis is expected because of matrix interference. However, the lower levels of mono-*ortho* PCBs in field samples after acid treatment was totally unexpected based on the literature reports which emphasize only the planar PCBs being affected by the matrix effects. Our results showed that the recovery of some mono-*ortho* PCBs was also affected by sample matrix which necessitated the reanalysis of the mono-*ortho* fractions besides the planar fractions. We, therefore, processed and measured PCBs in all the three cuts after column chromatography instead of only in the planar fraction as originally proposed to assess the contaminant levels. Because all the available resources had to be diverted to these unexpected additional analyses, PCNs (i.e., tetra and pentachloro-naphthalenes) in field samples could not be measured as originally planned. However this measurement will be pursued in the near future when new funds are available.

Results and discussion

Temporal distribution of PCB congeners in SM and SP Basin cores:

The concentration levels of the total, mono-*ortho*, coplanar and other PCBs (mostly di-*ortho*) in the core sections are presented in Table 1. The footnote presents the list of PCBs quantitated. The total PCBs vary from 3 to 273 ng/g and is uniformly less than the total PCBs reported earlier when the coplanar PCBs were not isolated by carbon column and hence was overestimated from matrix interferences (Venkatesan, 1998). The current values are generally less than the total PCBs reported for San Pedro Bay (450 ng/g) and Santa Monica Bay (720 ng/g) sediments, Palos Verdes Shelf (980 ng/g; NOAA, 1991) and several west coast bay and harbor sediments but greater than other areas throughout coastal United States (NOAA, 1988). Core IV adjacent to JWPCP outfalls contains the highest PCBs content and Core V which is also in San Pedro Basin, but farther offshore in deeper waters shows the second highest and Core I, in SM Basin, the third highest content of PCBs. Concentrations of total PCBs between 50 to 250ng/g are common in these sites which are potentially toxic concentrations (Safe, 1990) and could be toxic to marine life. Cores II and VI contain uniformly lowest levels among the cores studied. This general trend is concordant with that observed for pesticides suggesting a common source for PCBs and pesticides, the major source being sewage outfall (Venkatesan, 1998; Zeng and Venkatesan, 1999). “Other” PCBs (di-*ortho* and a few others, see Table 1 for explanation) dominate at the level of 59-92% of the total content. Specifically, coplanar PCBs which are considered to be the most toxic of the PCBs occur at very low levels and constitute the smallest percentage of the total. Mono-*ortho* PCBs which are believed to be next most toxic PCBs are found in all the cores in the mid-range (Table 1). BZ126 which is also a coplanar PCB was not detected in any of the samples even after carbon column procedure.

PCBs were manufactured in USA from 1929 to 1977 and were sold under the trade name Aroclor as a mixture of specific congeners. Because of their low flammability and high stability, PCBs have been utilized in transformers and capacitors, hydraulic fluids, lubricants, plasticizers, dyes, etc. (U.S. EPA, 1984). PCBs were in large use especially in the early 1970s and a ban on their production was enforced in 1977. Figure 3 illustrates the core profiles of PCB distribution in the SM/SP basins. Error bars are not shown because the values are too small for the scale.

The common use of PCBs apparently began to escalate from ~1940s peaking around the 1970s after which a decline in their use is noted, as evident specifically in Cores IV and V. This trend is similar for both total and selected (toxic) PCBs. Previously, Hom et al. (1974) analyzed for PCBs in a dated core from anoxic sediments of Santa Barbara basin in 1970. They observed a steady increase in PCBs content from ~1945 through 1967 and no recent core study has been undertaken in this site to document the recent decline in their concentration concomitant with the source control. Surface sediment grabs and short cores have been investigated by Stull et al. (1988) along the Palos Verdes shelf where the core profiles exhibit an increase in PCB content from the late 1930s and reach a maximum in the 1960s and 70s and a subsequent decrease in the recent times of PCBs content. An analogous trend in our samples consistent with the historic use of PCBs is clearly evident from Figure 3, especially from cores I, II, IV and V. Although PCB mass emissions from JWPCP outfalls has decreased by more than 99% from ~1300 kg in 1973 to ~80 kg in 1985 and below detection in 1988 (SCCWRP, 1989), the rate of decline of PCBs in surface sediments off Palos Verdes is not proportional to this decreased inputs (Mearns et al., 1991). Concentration of PCBs in the surface stratum in many cores is not trivial. Apparently, there is a significant increase in PCBs content in the surface section in Core IV corresponding to 1990-91. It is not clear if this elevated surface level can be fully justified by sediment mixing and this calls for additional sources of PCBs. Total PCB levels from <1 µg/g to as high as 11 µg/g dry weight of sediments have been reported in the Los Angeles Harbor which could be another significant land-derived runoff source of PCBs in Palos Verdes Shelf, (Eganhouse et al., 1990). This could also contribute to the wide distribution of PCBs in the water column off Palos Verdes Shelf (Zeng et al., 1999).

Preferential removal of *meta* and *para* chlorines (coplanar PCBs) were noted earlier in some studies of aquatic sediments and laboratory degradation (Brown et al., 1987; Quensen et al., 1988; Abramowicz, 1990). The main objective of our study was to identify if there is any such preferential degradation of specific PCBs, especially the most toxic coplanar PCBs and the toxic mono-*ortho* PCBs, with time, which should be evident from the historic profiles of the cores assuming the composition of the congeners in the total PCBs source remained nearly the same over the study period. There is no known record to indicate specifically that the formulation changed during this time either. The percentages in total PCBs of mono-*ortho*, coplanar and other PCBs (which includes di-*ortho* PCBs among others) as shown from representative examples in Figures 4 and 5 illustrate that there is no significant preferential degradation of any of the three groups of PCB congeners with depth. The shelf sediments are oxidizing and are not conducive for reductive dechlorination of the PCBs. However, in the anoxic basin sediments if coplanar PCBs were selectively degraded by reductive dechlorination as expected from previous reports, their percentage in total PCBs would decline with depth much more than what is concomitant with the source inputs. Instead, only a slight variation in % decrease in mono-*ortho* and coplanar PCBs with depth in some cores is noted and the general trend is an uniformly near constant percentage of the three groups of PCB congeners with increasing depth. Further, concomitant with reductive dechlorination, PCBs with lower chlorine numbers would have been generated. However, we did not find increase in the lower chlorinated *ortho* substituted PCBs like BZ8 or BZ28 with depth in any of the cores either. This is contrary to that reported in other studies (Brown et al., 1987; Quensen et al., 1988; Abramowicz, 1990).

The results also suggest that the even if degradation is occurring, it is not at a perceivable level in the time horizon represented by the cores and/or there is no significant /measurable bias in the diagenetic/microbial degradation among the three groups of PCB congeners in the sample

cores studied from the S. California Bight. This would, in turn, indicate that detoxification in the historic deposits analyzed here may not be occurring. However, all our samples had uniformly much lower levels of coplanar PCBs although some sections in cores I, IV and V contain more coplanar PCBs than others (Table 1, Figures 4 and 5). It is likely that degradation would have occurred if their levels were higher than that measured here. In general, the near constant and very low relative ratios of non-*ortho* to total PCBs in the strata in the cores probably indicate that the relative toxicity imparted by the coplanar PCBs is also probably similar in all the cores. The overall toxicity in the stratum is determined rather by the concentration of the total PCBs in their entirety.

Spatial distribution of PCB congeners in the Santa Monica Bay:

The total PCBs in the bay sediments (Figure 6, Table 2) range from 11 to 116ng/g dry sediment with the highest values around the 7-mile outfalls and the next highest in the southeast corner of the Bay, just north of the Redondo Canyon at stations E9, E10 and B10. This is consistent with our earlier study on DDTs from the Bay (Venkatesan, 2002). The remainder of the bay has PCB levels around 11-30ng/g. Mono-*ortho* PCBs constitute 11% to 27% of the total PCBs while coplanar PCBs are at very low levels ranging from 0.2 to 1% of the total with the exception of station C1 which has a higher percentage of 4% (Figures 7 & 8). The major portion of the total PCBs comprises other PCBs including di-*ortho* PCBs listed in Table 1. Figures 9 and 10 illustrate the distribution of coplanar and other PCBs respectively in the sediments arbitrarily grouped based on the water depth. The uniformity in their relative amounts (coplanar ~1% and others 75% -91% of the total PCBs with a very few exceptions) between stations at different water depths suggests that the PCBs are transported throughout the entire Bay with different sediment grain size with no significant bias to the various PCB congeners. This means that the toxic coplanar/mono-*ortho* PCBs do not preferentially accumulate in specific provenance within the Bay.

The total PCBs, however, appear to accumulate north of Redondo Canyon with fine grained sediments. Although this could have originated from the Hyperion discharges in the past, part of the PCBs could also have been transported from Palos Verdes Shelf via prevailing subsurface currents (California Countercurrent) advecting effluent particles upcoast along the bathymetric contours (Hickey, 1993; Stull et al., 1996). This scenario is also consistent with the modeling studies of Niedoroda et al.(1996) who reported that the contaminated particles from the major historic deposits in the Palos Verdes Shelf are dispersed upward by bioturbation and then removed by strong flow events to wider marine environment including SM Bay to the northwest. In summary, PCBs are found widespread in the Bay at different levels with hot spots of relatively higher concentration either from their proximity to the source and/or from transport of material from Palos Verdes Shelf into the SM Bay where conditions are conducive to the accumulation of organic matter.

Conclusions

The core profiles of distribution of PCBs document the impact of the change in wastewater technology in the region. No significant preferential degradation of any of the three groups of PCB congeners (coplanar, mono-*ortho* and di-*ortho*) with depth in the SM/SP sediment cores was observed. Specifically, no measurable reductive dechlorination and resultant detoxification of the coplanar PCBs has occurred in the anoxic basin sediments in the last several decades. It appears that almost all of the PCB congeners are relatively refractory in the sediment

cores investigated. The near constant and very low relative ratios of non-*ortho* to total PCBs in the strata in the cores probably indicate that the relative toxicity imparted by the coplanar PCBs may be similar in all the cores. The overall toxicity in the stratum is determined rather by the concentration of the total PCBs in their entirety.

PCBs are found widespread in the Santa Monica Bay surface sediments with hot spots of relatively higher concentrations near the outfalls pipes and in the southeast corner of the Bay, just north of Redondo Canyon. The results also indicate that there is no preferential accumulation of coplanar and mono-*ortho* PCBs over di-*ortho* congeners in specific provenance within the Bay.

This study in conjunction with sediment dynamics data should, therefore, be useful to the LA City and County Sanitation Districts, aquaculture industry and the public which use and enjoy the coastal waters. The results should serve as a good scientific basis for historic deposits of contaminants and their fate in the coastal region for the State Water Resources Board, when they are ready to develop “sediment quality objectives”.



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Cooperating Organizations

City of Los Angeles, Bureau of Sanitation

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Figure captions:

Figure 1. Locations of box core sediments.

Only five cores, I, II, IV, V and VI were analyzed. (Cores I-VI were collected for a NOAA/NS&T Program). 1 and 2 are dumpsites. H= Hyperion, City of Los Angeles, and C= Los Angeles County Sanitation Districts.

Figure 2. Locations of surface grab sediment samples from the Santa Monica Bay.

Open circles: 2001 collection

Open squares: 2001 and 2002 collection

E3, B6 and E10: 2000, 2001 and 2002 collection

Z2: 1999, 2000, 2001 and 2002 collection

Figure 3. Historic profiles of total PCBs in the box cores. Note: The values are plotted against the mid-point (i.e., 1.5 cm for a section from 1-2 cm) of each section in the core in this and all other figures (Fig. 4 and 5) on core data.

Figure 4. Percentage in total PCBs of mono-*ortho*, coplanar and di-*ortho* PCB congeners in core IV.

Figure 5. Percentage in total PCBs of coplanar PCB congeners in core V and VI and di-*ortho* congeners in core VI.

Figure 6. Total PCBs in the SMB sediments in ng/g dry sediment. Average values for multiple year collection plotted.

Figure 7. Percentage of mono-*ortho* PCB congeners in total PCBs in SMB sediments.

Figure 8. Percentage of coplanar PCB congeners in total PCBs in SMB sediments.

Figure 9. Percentage of coplanar PCB congeners in total PCBs in SMB sediment groups based on water depth.

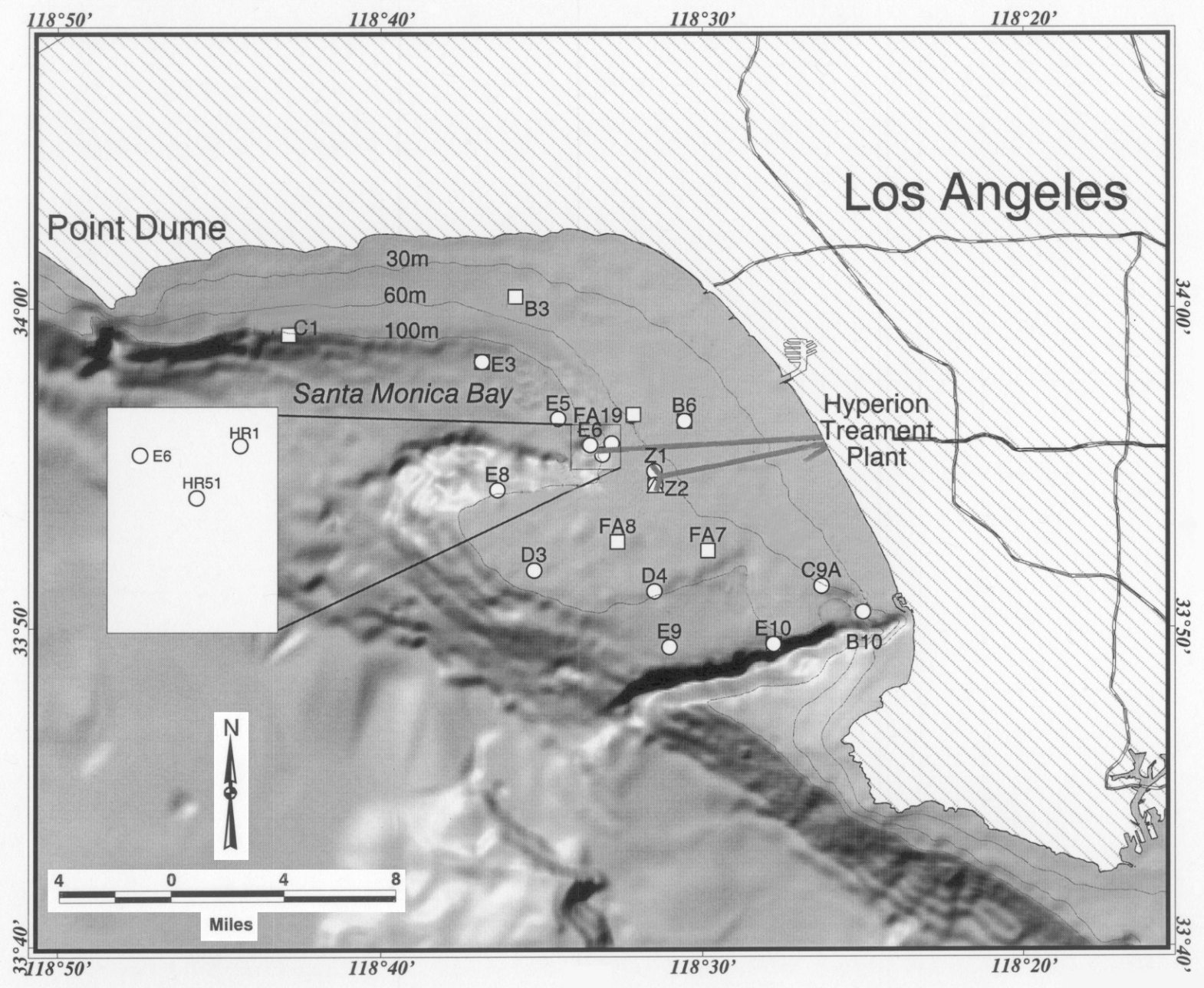
Figure 10. Percentage of other PCB congeners in total PCBs in SMB sediment groups based on water depth.

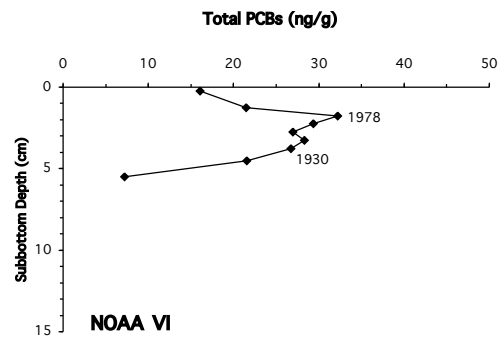
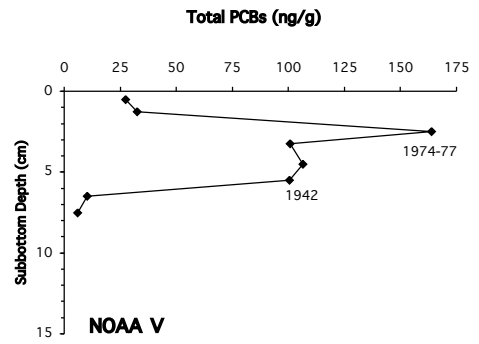
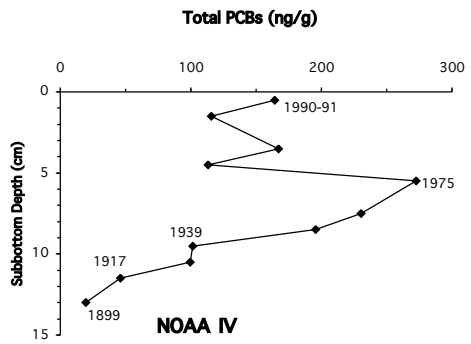
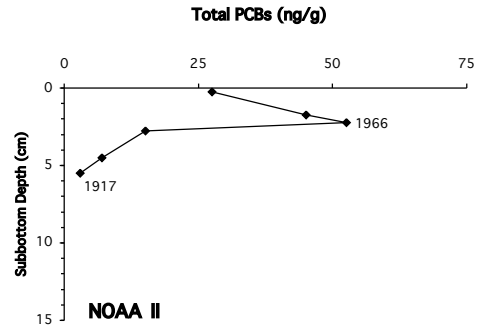
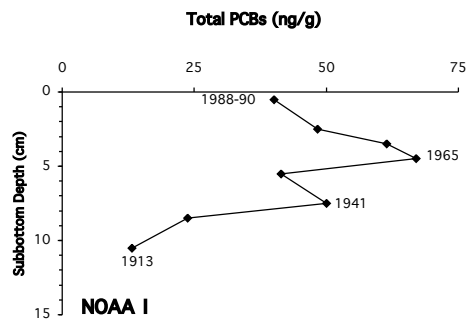
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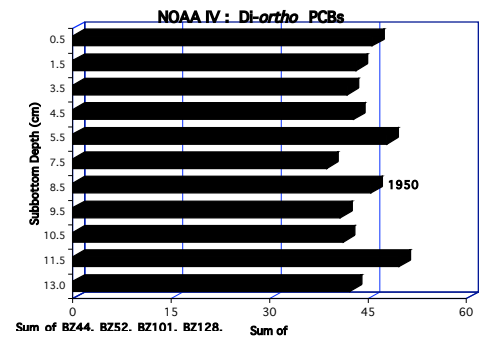
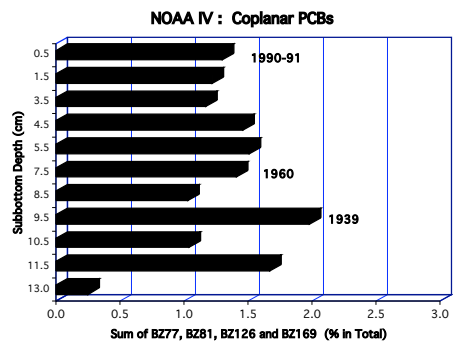
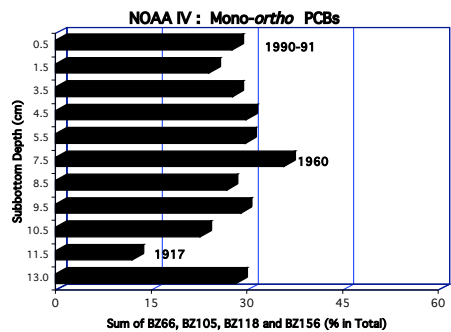
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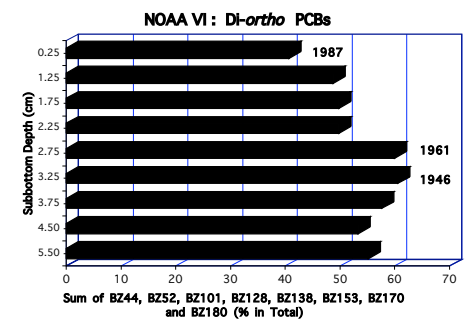
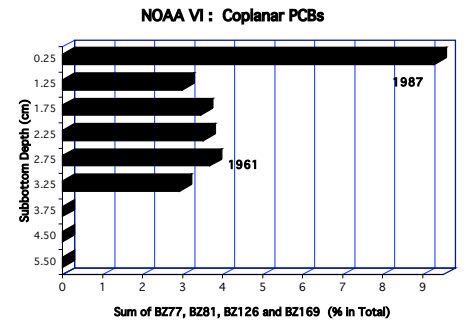
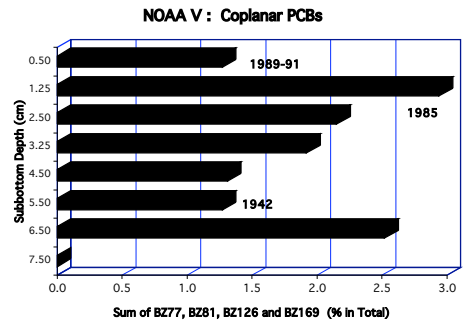
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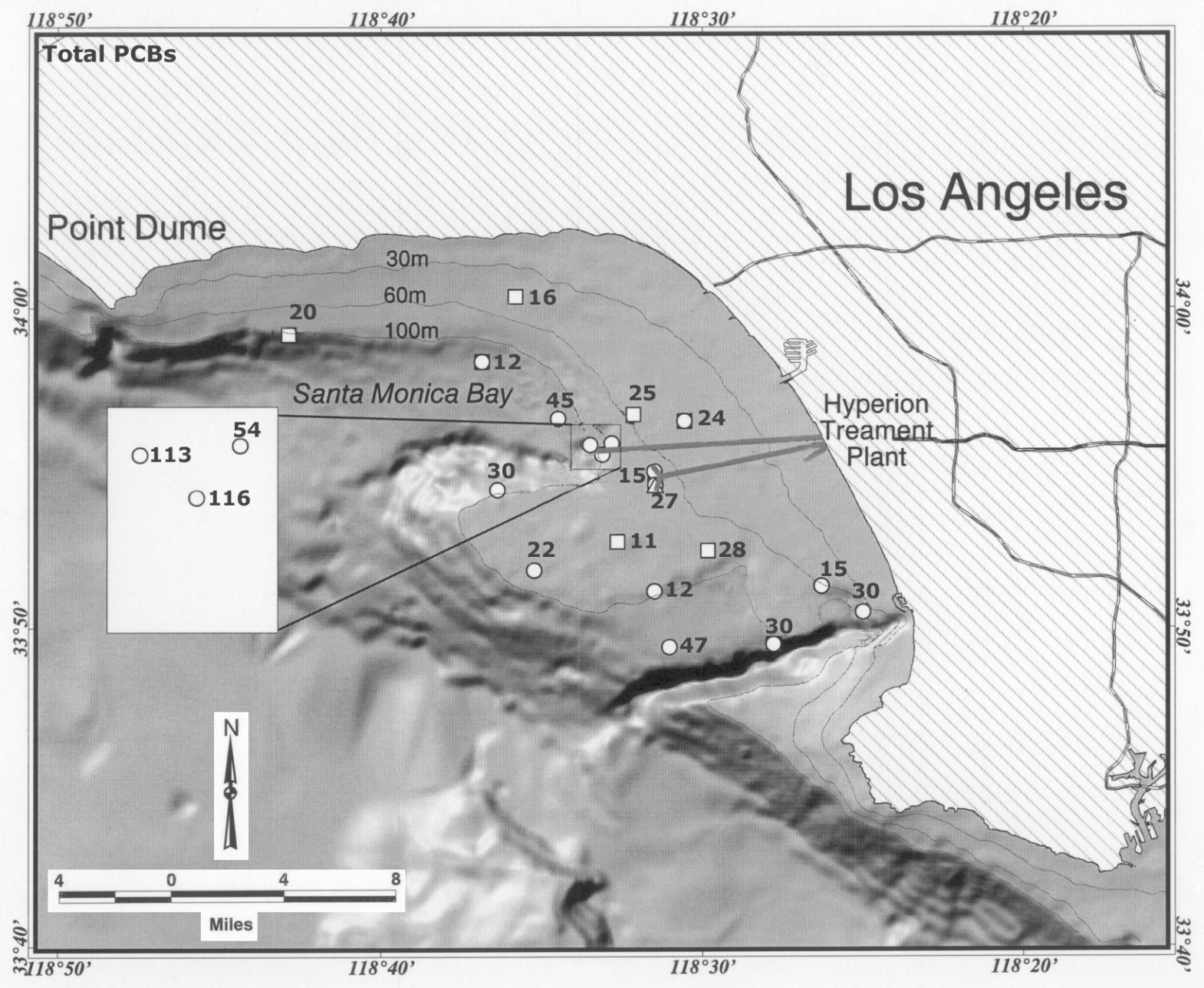


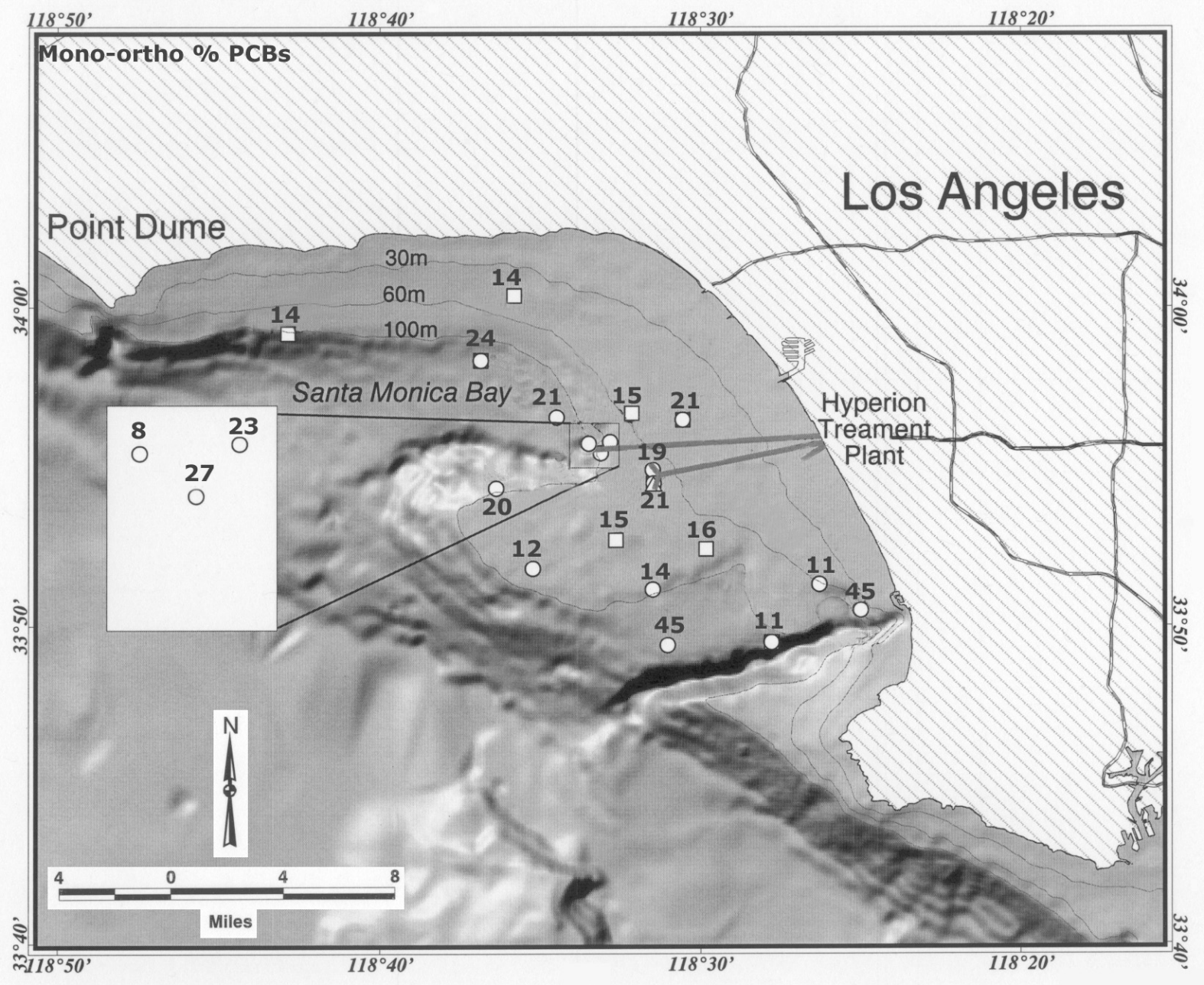


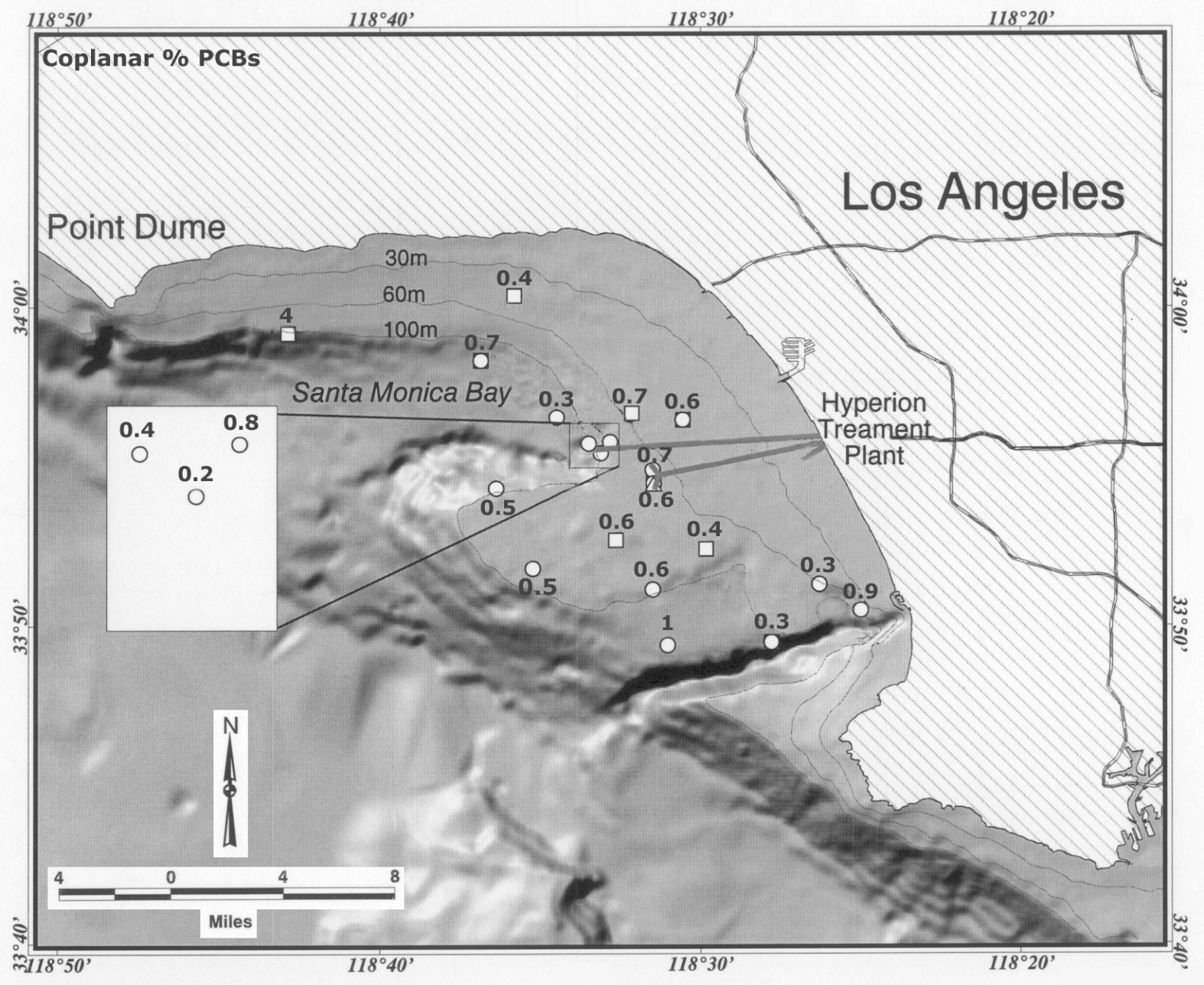




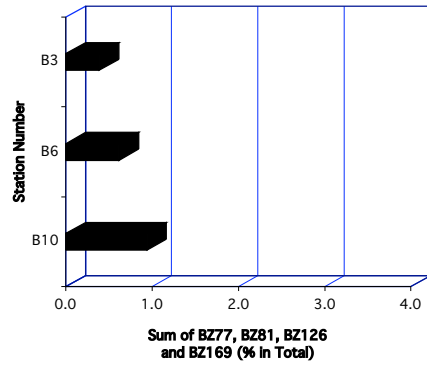




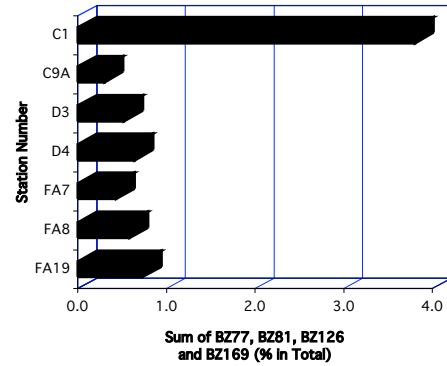




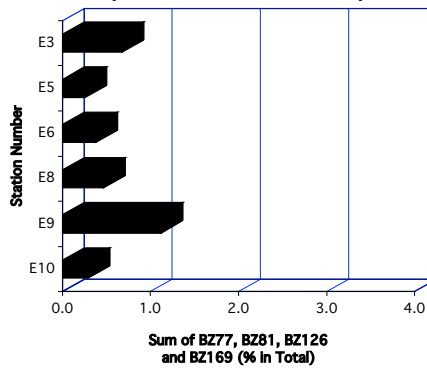
Coplanar PCBs at 45 m Water Depth



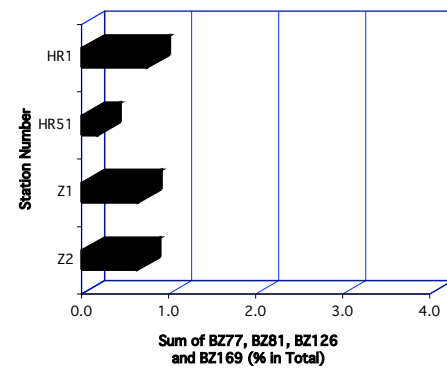
Coplanar PCBs at 60-80 m Water Depth



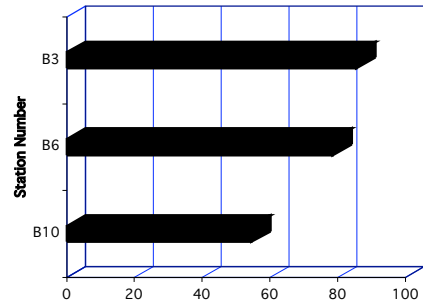
Coplanar PCBs at 150 m Water Depth



Coplanar PCBs at vicinity of Outfall pipes pipi

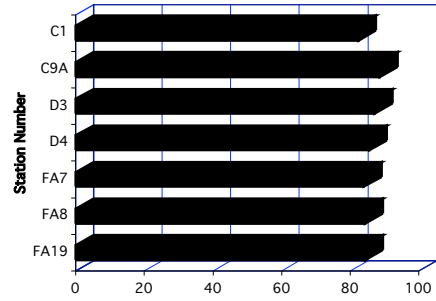


Other PCBs at 45 m Water Depth



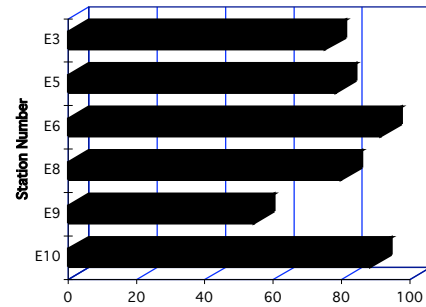
Sum of BZ8, BZ18, BZ28, BZ44, BZ52, BZ101, BZ128, BZ138, BZ153, BZ170, BZ180, BZ187, BZ195 and BZ206 (% in Total)

Other PCBs at 60-80 m Water Depth



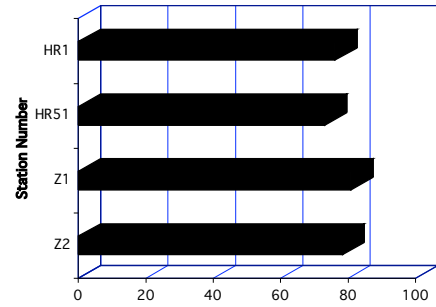
Sum of BZ8, BZ18, BZ28, BZ44, BZ52, BZ101, BZ128, BZ138, BZ153, BZ170, BZ180, BZ187, BZ195 and BZ206 (% in Total)

Other PCBs at 150 m Water Depth



Sum of BZ8, BZ18, BZ28, BZ44, BZ52, BZ101, BZ128, BZ138, BZ153, BZ170, BZ180, BZ187, BZ195 and BZ206 (% in Total)

Other PCBs at Vicinity of Outfall Pipes



Sum of BZ8, BZ18, BZ28, BZ44, BZ52, BZ101, BZ128, BZ138, BZ153, BZ170, BZ180, BZ187, BZ195 and BZ206 (% in Total)

Table 1. Polychlorinated biphenyls in Santa Monica and San Pedro Basin sediment cores

Station/Core	Mono-ortho*	Coplanar**	Other***	Total#
NOAA I				
Concentration(ng/g)	2-20	0.1-7	11-45	13-67
% in total PCBs	17-30	1-12	59-81	
NOAA II				
Concentration(ng/g)	0.05-8	0.2-1	3-45	3-53
% in total PCBs	2-17	1-6	79-92	
NOAA IV				
Concentration(ng/g)	6-83	0.05-4	14-186	19-273
% in total PCBs	12-36	<1-2	62-84	
NOAA V				
Concentration(ng/g)	0-27	0-4	4-130	6-164
% in total PCBs	0-21	0-3	70-90	
NOAA VI				
Concentration(ng/g)	1-11	0-1.5	6-23	7-32
% in total PCBs	12-27	0-4	61-85	

*Sum of BZ 66,105,118 and 156

**Sum of BZ 77, 81, 126 and 169. Note BZ 126 was not detected in any sample.

***Sum of BZ 8, 28, di-ortho PCBs (BZ 18, 44, 52, 101,153, 128, 180, 170, 138) and tri-ortho PCBs (BZ 187, 195 and 206).

#Sum of all of the above.

Table 2. Polychlorinated biphenyls in the Santa Monica Bay sediments

Station	Mono-ortho^a	Coplanar^b	Other^c	Total^d
Water depth 45 m*				
Concentration(ng/g)	2-14	<0.1-0.3	13-19	15-33
% in total PCBs	14-45	0.4-0.9	55-86	
Water depth 60-80 m**				
Concentration(ng/g)	2-4	<0.1-0.8	9-24	11-29
% in total PCBs	11-17	0.4-4	84-89	
Water Depth 150 m[#]				
Concentration(ng/g)	3-10(21 ^e)	<0.1-0.5	9-103	12-124
% in total PCBs	8-45	0.3-0.7	75-91(54)	
Vicinity of outfall pipes^{##}				
Concentration(ng/g)	6-31	0.1-0.4	12-85	18-116
% in total PCBs	19-28	0.2-0.8	73-81	

^aSum of BZ 66,105,118 and 156

^bSum of BZ 77, 81, 126 and 169. Note BZ 126 was not detected in any sample.

^cSum of BZ 8, 28, di-ortho PCBs (BZ 18, 44, 52, 101,153, 128, 180, 170, 138) and tri-ortho PCBs (BZ 187, 195 and 206).

^dSum of all of the above.

^eAbnormal value for the set is given in parenthesis.

*SM bay stations: B3, B6, B10

**SM Bay stations: C1, C9A, D3 D3, D4, FA7, FA8, FA19

[#]SM Bay stations: E3, E5, E6, E8, E9, E10

^{##}Stations at the outfall pipes: HR1, HR51, Z1, Z2