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Some Factors Affecting the Recovery of Polychlorinated Biphenyls (PCB's) from Water and Bottom Samples

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ABSTRACT: During studies on analytical methods for PCB's in water and bottom samples, variable recoveries from dosed samples and apparent decreasing recoveries with the age of the sample were observed. Losses from dosed river waters as high as 10 percent in one day and greater than 40 percent in one week were common. Recoveries from bottom samples varied widely with the sample pretreatment and the method of extraction. In an effort to define some of the factors affecting the recovery of PCB's from water, the following parameters and conditions were studied: sample container, extremes of pH, aging under ambient conditions, and several preservation techniques. Several methods of sample preparation and extraction of bottom samples were studied. Environmentally contaminated water and lake bottom samples and dosed natural samples were studied.

The separatory funnel liquid-liquid extraction method was found to be the most efficient for the extraction of PCB's from natural waters. The air-dried, moisture-added soxhlet extraction procedure proved to be the most efficient for recovery of PBC's from environmentally contaminated lake bottom samples. Formaldehyde was found to be effective for preserving PCB's in dosed natural waters for at least two weeks.

KEY WORDS: water quality, water pollution, pesticides, toxicity, recovery, aquatic animals, polychlorinated biphenyls (PCB's)

Polychlorinated biphenyls (PCB's) have been recognized for several years as ubiquitous environmental pollutants. These complex mixtures of variably chlorinated biphenyls have occurred in nearly every environmental media [*1,2*].[2] Their presence in water, sediment, biota, fish, birds, and animals have been reported [*3-5*]. While the PCB's are reported to be less toxic than the chlorinated hydrocarbon pesticides, acute toxic effects to aquatic life have been demonstrated [*3,6*]. Since they do exhibit acute

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[2] The italic numbers in brackets refer to the list of references appended to this paper.

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toxic effects and, like the chlorinated pesticides, they are biologically magnified [7], it is important to maintain surveillance of the PCB levels occurring in the environment.

The PCB's are chemically similar to many of the organochlorine pesticides; therefore, the same analytical procedures are applied to both. Being complex mixtures, the PCB's present a serious interference problem when determining many pesticides. Therefore, special separation and cleanup procedures are required for analyses when both occur in a given sample. Much work has been published on the separation and the determinative aspects of the analysis [*8-10*]. However, little has been reported on factors that might affect the extraction and recovery of PCB's from environmental samples.

During our studies on water, we noted variable recoveries from dosed samples and an apparent decreasing recovery with the age of the sample. In an effort to define some of the factors affecting the recovery, the following parameters were investigated: the sample container, extremes of pH, aging under ambient conditions, and several preservation techniques. Liquid-liquid extraction and extraction with urethane foam plugs were studied. During our studies on recovery of PCB's from bottom samples, several methods for preparation and extraction of bottom samples were compared. We were particularly interested in the newer column extraction procedures since they appear to be becoming very popular. Environmentally contaminated samples and dosed natural samples were studied.

Experimental

Apparatus—The high frequency mechanical dispersion instrument used in one phase of the bottom sample extraction study was the Super Dispax Tissumizer, Model STD 182N (Tekmar Company, Cincinnati, Ohio).[3]

Reagents—Reference Standard PCB's Aroclors 1242, 1254, 1260, and 1016 (Monsanto Chemical Company, St. Louis, Mo.) prepared in acetone solution. Formaldehyde—37% aqueous solution.

Procedure—The samples were collected in 1-qt to 6-gal glass containers at various times over a period of one year.

The primary liquid-liquid extraction procedure employed for water samples was that recommended by the U.S. Environmental Protection Agency (EPA) for organochlorine pesticides [*11*]. In addition, the semi-automatic magnetic stirring (Vortex) system of Kawahara et al [*12*] was tested. The urethane foam procedure for extraction of PCB's from water was that published by Gesser et al [*13*]. A modification of Gesser's procedure, that of soaking the foam plugs in the sample with occasional shaking, was also tested.

[3]Mention of products and manufacturers is for identification only and does not imply endorsement by the Methods Development and Quality Assurance Research Laboratory of the U.S. Environmental Protection Agency.

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Several methods for extraction of bottom materials were investigated. The soxhlet extraction procedure of Breidenbach et al [*14*], column elution procedures of Hesselberg and Johnson [*15*] and Southeast Environmental Research Laboratory [*16*], the high frequency dispersion procedure of Johnson and Starr [*17*], the mechanical shaking procedure of Goerlitz and Brown [*18*] and Shell Chemical Company [*19*], and the blender extraction method of Muth [*20*] were tested.

All bottom sample extracts were cleaned up in the same manner. The standard Florisil column procedure [*11*] and the semi-micro silica gel procedure of Leoni [*9*] were employed. When necessary, elemental sulfur was removed from the extracts using mercury according to the method of Goerlitz and Law [*21*]. Table 1 lists the conditions for preparation and extraction of bottom samples.

TABLE 1—*Extraction procedures tested for recovery of PCB's from bottom samples.*

Method	Ref	Preliminary Drying	Dry Matter, %	Water Added	Extracting Solvent
Soxhlet	[*14*]	air dry at room temperature, grind—mortar and pestle	>95	10%	hexane-acetone (9:1)
Shake I	[*19*]	air dry at room temperature, grind—mortar and pestle	>95	slurry	hexane-isopropyl alcohol (3:1)
Shake II	[*18*]	none	...	none	acetone with hexane added later
Blender	[*20*]	Na_2SO_4 added		none	acetonitrile-acetone (1:1)
Column I	[*15*]	partial air dry at room temperature, mix with Na_2SO_4 and grind	~70	none	hexane
Column II	[*16*]	partial air dry at room temperature, mix with Na_2SO_4 and grind	~50-70	none	hexane-acetone (1:1)
High frequency dispersion	[*17*]	none	...	none	acetone

Dosing Procedures—Water Samples (880 ml) in 1-qt sample bottles were individually dosed by pipetting the PCB's in acetone solution (1 μg/ml) and shaking vigorously to achieve uniform mixing.

Bottom Samples—Approximately 400 g of an air dried sediment (98.7 percent dry matter) was ground with a mortar and pestle and passed through a 20 mesh screen. The sediment was then placed in a 1-qt wide mouth glass bottle. Distilled water (200 ml) was added and mixed to form a slurry. Aroclor 1242 (50.0 μg) and Aroclor 1254 (50.0 μg) in 1 μg/μl

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acetone solution was added to the slurry with continual mixing. The bottle was sealed and allowed to equilibrate for 20 h with occasional shaking. Appropriate aliquots of the dosed sample were taken and treated as required by the extraction methods to be tested. The high frequency dispersion device, Column II, and soxhlet extraction were selected for testing recovery on dosed bottom samples.

Quantitation Techniques—Two methods were used to reduce the electron capture gas chromatographic data:

(*a*) When a dosed or environmentally occurring PCB equivalent to or closely resembling a standard Aroclor was present, a simple calculation was made by summing all the peak heights of the unknown and comparing it to the sum of the peak heights of the reference Aroclor.

(*b*) When mixtures of PCB's, not representing a single Aroclor were present, the method of Webb [*10*] was used to determine the PCB concentrations and the most likely Aroclor represented.

All results for bottom samples were calcualted on a dry weight basis. The dry weight was obtained by oven drying an aliquot of the sample at approximately 105°C overnight.

Gas chromatography and mass spectrometry techniques were used to confirm the identity of the PCB's in the environmentally contaminated samples.

Results and Discussion

Water Samples

Initial studies to determine the cause of low and variable recoveries of PCB's from dosed natural waters centered around the bottle cap liners. Teflon lined caps which are used routinely are considered the best choice when collecting samples for organic analysis. It was thought that the Teflon may be sorbing some of the PCB's. However, no PCB's were recovered from Teflon liners after soaking in hexane for 1 h. Subsequently, a comparison of recoveries using Teflon lined caps and aluminum lined caps was made. The samples were stored under ambient conditions in the laboratory, up to 11 days. Figure 1 shows that there is a tendency toward higher recoveries when using aluminum lined caps. In each case, samples stored with aluminum cap liners yielded greater recoveries than those stored with Teflon lined caps.

Sample Preservation

Figure 2 shows the results of aging on recovery of PCB's from dosed natural river waters. Samples held at ambient conditions in the light showed variable and often low recoveries with a general decrease in

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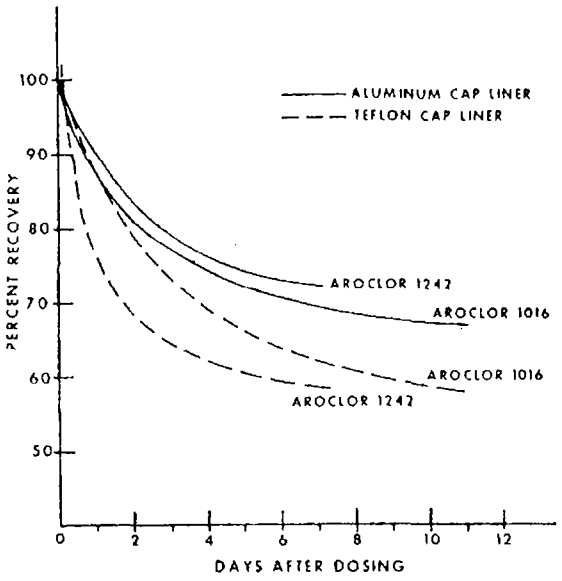

FIG. 1—*Effect of cap liner on recovery of PCB's from dosed river water (dose of 1 μg per sample).*

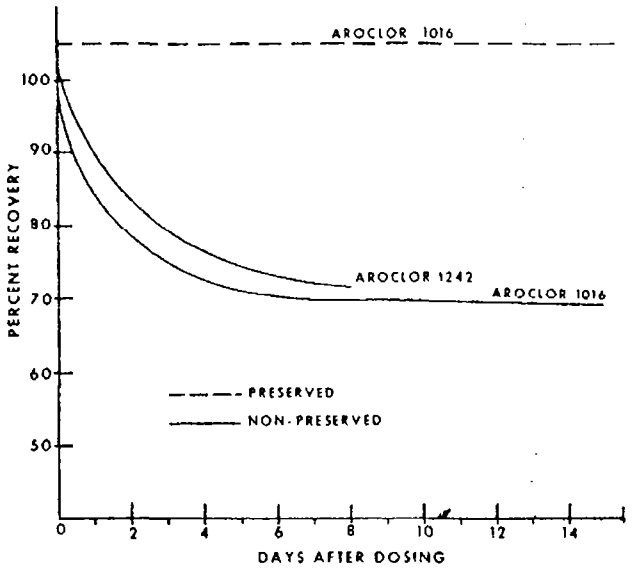

FIG. 2—*Effect of aging on recovery of PCB's from dosed river water at ambient temperature, preserved versus non-preserved (dose of 1 μg per sample).*

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recovery with time. Recoveries ranged from 102 percent to a low of 67 percent over a period of 15 days. The recoveries from samples preserved with formaldehyde were quantitative over the time period, averaging 105 percent and ranging from 92 to 116 percent. Figure 3 depicts typical losses of the isomers of Aroclor 1016 that occur after aging a dosed sample for 15 days. The greater losses occur in the isomers that contain three or fewer chlorine atoms. The greatest losses occur in peaks 37 and 40 which represent PCB's containing three chlorine atoms. All natural water samples tested showed a trend toward reduced recovery with the age of the sample. The rate of loss appeared to be affected by the variation in physical, chemical, and biological characteristics of the samples.

Table 2 compares the recoveries of Aroclor 1016 obtained from dosed river waters that have been preserved with formaldehyde up to 15 days to the recoveries obtained at time zero (<8 h after dosing). A statistical

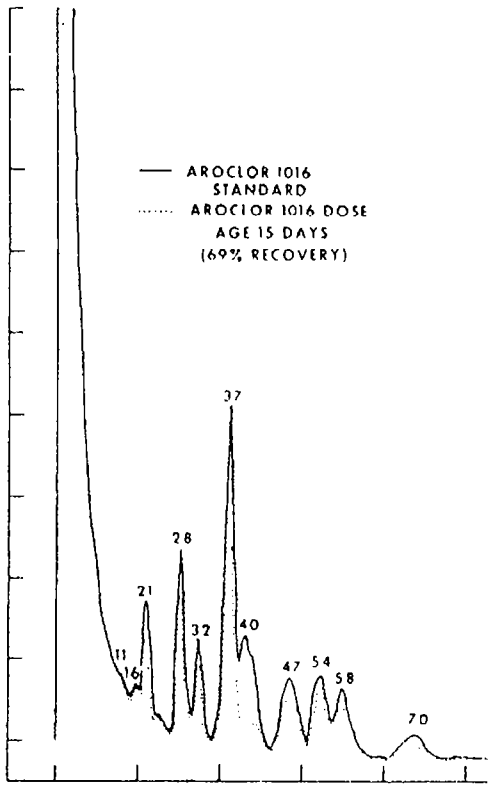

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FIG. 3—*Electron capture gas chromatogram of reference standard and recovered Aroclor 1016.*

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TABLE 2—*Recovery of Aroclor 1016 from river waters at time zero versus samples preserved and aged* (dose, 1 μg *sample*).

	Recovery, %	
	Time Zero[a]	Preserved[b]
	109	92
	107	107
	107	110
	98	111
	107	109
	116	93
	111	116
	102	111
	92	104
	109	105
	111	108
	100	111
	99	95
	105	
Mean	105.4	105.5
No. of samples	14	13
Standard deviation	6.4	7.6
Variance	40.4	57.8
Coefficient of variation	6.0	7.2
Blank value (average)	6.0	6.0
Corrected mean	99.4	99.5

[a] Time zero = <8 h after dosing.
[b] Preserved with formaldehyde (15 ml of 37% solution).

comparison of the data shows very good agreement between the time zero and preserved samples.

The pH of the sample had no effect on the extraction efficiency from dosed samples over the range of 2.7 to 10.5. That is, the average recovery at time zero for the pH levels listed in Table 3 was quantitative. The accuracy and precision was essentially equivalent to that obtained for the untreated samples shown in Table 2. The limited data obtained suggest an apparent effect on the recovery as the sample ages at pH 5.3 and 7.3. Recoveries at these pH levels decreased with time although the decrease was not as great as noted in other studies. Recoveries at pH 2.7, 8.8, and 10.5 were near quantitative throughout the time period studied.

The effect of storing samples at ambient temperatures in the light, dark, and under refrigeration was studied. The results are listed in Fig. 4 and compared to results obtained for samples preserved with formaldehyde. Samples stored in the dark yielded higher recoveries throughout the period of study than those stored in the light. Refrigerated samples gave slightly better recoveries than those stored at ambient temperature in

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TABLE 3—*Recovery of Aroclor 1016 from dosed river water at various pH levels (dose, 1 μg/sample).*

pH	Age, days	Recovery, %
2.7	2	98
2.7	6	102
2.7	14	91
5.3	2	107
5.3	6	89
5.3	14	86
7.3	2	109
7.3	6	87
7.3	14	84
8.8	2	107
8.8	6	106
8.8	14	95
10.5	2	107
10.5	6	105
10.5	14	100

NOTE—
Blank value = 7%.
Day zero value (mean recovery) = 107%.

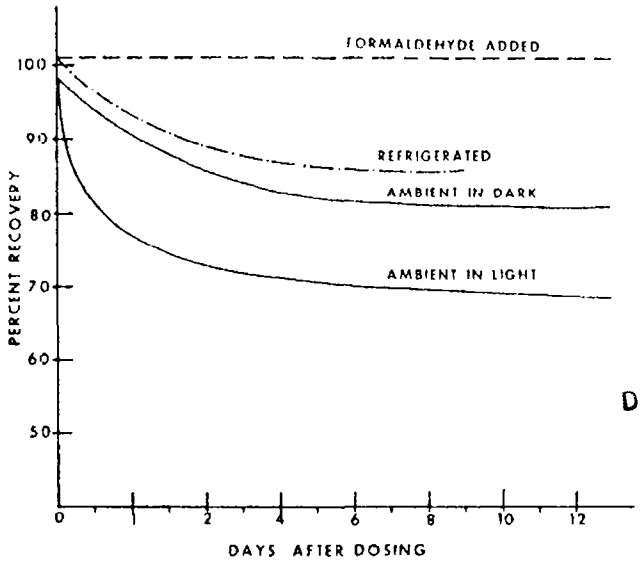

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FIG. 4—*Recovery of Aroclor 1016 from dosed river water when stored at ambient temperature (in light and the dark), under refrigeration, and preserved with formaldehyde (dose of 1 μg per sample).*

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the dark. However, the trend of decreasing recovery was evident under all three conditions. Samples preserved with formaldehyde gave the highest and least variable recoveries and showed no decreasing trend in recovery with the age of the sample.

The use of the semi-automatic liquid-liquid extraction system of Kawahara et al [*12*] was considered. A preliminary evaluation showed it to be inferior to the separatory funnel shake out. Therefore, it was not tested further. At time zero, the mean recovery using this system was 84 percent with a range of of 77 to 92 percent.

Polyurethane Foams

The use of urethane foam plugs as reported by Gesser et al [*13*] appeared to show promise as a means for concentrating and isolating PCB's. Thus, we evaluated the technique on dosed distilled and natural river water. Table 4 lists the data obtained using the column procedure of Gesser and by soaking the plugs in the sample. The column method worked well on distilled water and was more efficient than the soaking procedure. However, the column method was not at all suitable for use on

TABLE 4—*Recovery of Aroclor 1016 from distilled water using polyurethane foam plugs (dose, 1 μg/sample).*

	Recovery, %	
	Column Elution	Soaking
	84	61
	79	89
	100	77
	95	58
	91	77
	71	95
	109	106
	91	109
	100	86
	86	50
	98	77
	76	50
	91	97
	88	60
	94	75
	95	114
	84	66
	99	98
	114	99
		100
Mean	91.8	82.2
Number of samples	19	20
Standard deviation	10.7	20.0
Variance	114.6	399.0
Coefficient of variation	11.6	24.3

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natural waters. Suspended material in the samples plugged the column. We were unable to pass the total volume of a 1-liter natural river water sample through the column without adding external pressure, in which case channeling occurred. Soaking the plugs in dosed river water worked, however, the accuracy and precision was poorer than that obtained with distilled water. Coating the plugs with SE-30 did not improve the efficiency. These results coupled with the high background material from the foam which had significant adverse effects on the gas chromatographic (GC) column and the detector were the bases for rejecting the method.

As a result of the foregoing studies, the separatory funnel liquid-liquid extraction technique was shown to be the most efficient for recovery of PCB's from natural waters. Therefore, this method was used for analyzing all wastewater samples described next.

Wastewater samples

Several industrial wastewater samples were collected and analyzed for environmental levels of PCB's. In one case, additional aliquots of the sample were then dosed with 5 μg/liter of Aroclor 1242 and again analyzed. Table 5 presents the results and statistical data derived from the

TABLE 5—*Recovery of PCB's from environmentally contaminated industrial waste samples. concentration of Aroclor 1242* (μg liter).

Replicates	870	870[a]	832	833
1	2.17	7.99	58.7	13.7
2	1.92	7.74	63.0	13.8
3	1.86	7.42	64.8	
4	2.27	6.97	66.9	
5	1.86	6.42	60.3	
6	1.70	6.79	66.3	
7	1.73	6.75	61.7	
Mean	1.93	7.15	63.1	
No. of samples	7	7	7	
Standard deviation	0.214	0.575	3.07	
Variance	0.046	0.330	9.46	
Coefficient of variation	11.11	8.03	4.87	

[a] Dosed with 5 μg liter of Aroclor 1242.

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samples. The results show good precision for two levels of environmental contamination. The precision for the dosed samples was equally good. The recovery for the dosed sample averaged 103 percent.

Bottom Samples

Bottom samples collected from a large man-made lake in Ohio were found to be contaminated with Aroclor 1242 and 1254. Samples collected from three locations in the lake were selected in order to evaluate seven

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extraction procedures. The total quantity of sample available to us limited the numbers of analyses that could be made. Nevertheless, useful information was derived from the results. Table 6 lists the results of our study on the lake bottom samples. The mechanical dispersion apparatus was not available at the time and no comparison of this technique could be made

TABLE 6—*Recovery of PCB's from environmentally contaminated lake bottom samples.*

Method	Ref	Sample[a] Area	Dry Matter, %	Aroclor 1242, μg/kg	Recovery,[b] %	Aroclor 1254, μg/kg	Recovery[b] %
Soxhlet	[14]	3	96.7[c]	26.2	100	12.5	100
Shake I	[19]	3	96.7	15.3	58.4	7.4	59.2
Blender	[20]	3	37.3	5.6	21.4	9.4	75.2
Shake II	[18]	3	37.3	1.0	3.8	2.7	21.6
Soxhlet	[14]	5	96.4[c]	54.3	100	24.7	100
Column I	[15]	5	76.9	6.4	11.8	8.9	36.0
Soxhlet	[14]	8	98.4[c]	0	. . .	403	100
Shake I	[19]	8	98.4	0	. . .	377	93.5
Column I	[15]	8	98.4	0	. . .	343	85.1
Column I	[15]	8	70.2	0	. . .	318	78.9
Blender	[20]	8	70.2	0	. . .	343	85.1
Blender	[20]	8	56.0	0	. . .	246	61.0
Shake II	[18]	8	70.2	0	. . .	321	79.7
Shake II	[18]	8	56.0	0	. . .	258	64.0

[a] Samples from Areas 3 and 5 were muck; samples from Area 8 were sandy muck.
[b] Recovery for soxhlet procedure considered to be 100%.
[c] 10% water added prior to extraction.

on these samples. The results obtained show that the air-dried, 10 percent moisture-added soxhlet procedure provides significantly greater efficiency than any of the other methods tested.

In addition, a study of the recovery of Aroclor 1242 and 1254 from dosed bottom samples was carried out using three methods—mechanical dispersion, column II, and the soxhlet. Results of this test (Table 7) show the superiority of the air-dried moisture-added soxhlet extraction pro-

TABLE 7—*Recovery of PCB's from dosed bottom samples (dose 237 μg/kg total PCB's).*[a]

Method	Ref	Dry Matter, %	μg Recovered		Average Recovery, %
			1	2	
High frequency dispersion	[17]	52.5	[b]	210	88.7
Column II	[16]	89.7	208	187	83.5
Soxhlet	[14]	98.4	230	226	96.2

[a] Mixture of Aroclors 1242 and 1254, 118.5 μg/kg of each.
[b] Sample lost due to malfunction of dispersion apparatus.

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cedure over the other extraction procedures as far as bottom sediments are concerned.

We acquired a sewage treatment plant sludge which was found to contain PCB's. Aroclors 1242 and 1260 were identified in the sample. Two extraction techniques were employed in the analysis—mechanical dispersion and soxhlet extraction. The results are listed in Table 8. In this case, equivalent results were obtained with the two methods.

TABLE 8—*Recovery of PCB's from environmentally contaminated treatment plant sludge.*

			Aroclor Recovered (mg/kg)	
Sample No.	Method	Dry Matter, %	1242	1260
831-A	high frequency dispersion	18.1	34.4	14.2
831-B	soxhlet	93.2	34.4	12.5

The results of these studies show the air-dried, 10 percent moisture-added soxhlet extraction procedure to be the most efficient method for recovery of PCB's from bottom samples. This is consistent with the results reported by others for recovery of certain organochlorine pesticides from soils [*22-25*]. The better efficiency of this procedure is probably due to the smaller particle size achieved when pulverizing the dry sediment and to the longer solvent contact time provided by the soxhlet method as reported by Saha et al [*26*] and Chiba and Morley [*27*]. Nash et al [*28*] evaluated soxhlet, shake, and column procedures somewhat different from those that we studied and concluded that his soxhlet and shake procedures gave near equivalent recoveries for nine pesticides from soil. In this case, the samples were not air dried or ground with a mortar and pestle prior to soxhlet extraction.

The cleanup procedures employed, the standard Florisil and the micro silica gel columns, were adequate for all industrial waste and sludge samples. However, all bottom samples required further treatment with mercury to remove sulfur. All procedures which required a water wash of the extracts, the shake, Column H, blender, and the high frequency dispersion methods, produced severe emulsions and additional manipulative steps not required by the soxhlet procedure.

The high frequency dispersion technique looks promising, at least, for sludge samples and should be evaluated further. However, some problems have been encountered in its use. Small stones, pea size and even smaller, are a hazard. They are thrown at terrific speeds into the wall of the container and will shatter glassware. Further, even small particle size sand causes excessive wear on the stator and rotor, in which case frequent parts replacement is required.

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Conclusions

The separatory funnel liquid-liquid extraction procedure is the method of choice for recovery of PCB's from natural waters. The air-dried soxhlet extraction procedure is the method of choice for recovery of PCB's from bottom and sludge samples. Formaldehyde effectively preserves PCB's in dosed natural water samples, at least up to two weeks.

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