

## Polychlorinated Biphenyl, Pesticide, and Heavy Metal Residues in Swine Foraging on Sewage Sludge Amended Soils

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Berkshire sows were overwintered for two seasons on experimental plots which had received various applications of Chicago sewage sludge for the 8 preceding years. Cadmium was the only element analyzed which accumulated significantly in tissues; all three levels of treatment were adequate to induce small amounts of Cd binding protein in livers and kidneys. Polychlorinated biphenyls and *p,p'*-DDE accumulated in the fat of the sows in a dose-related manner. Individual chlorobiphenyls were analyzed, and the results indicate that estimation of Aroclor content by measuring selected peaks results in misleading information.

The use of municipal sewage sludge as a soil amendment is practiced in several areas of the world in order to alleviate a burdensome disposal problem while reclaiming a valuable resource. In the United States, the practice is becoming highly regulated because of concerns about the accumulation of heavy metals and persistent organic chemicals (*Fed. Regist.*, 1979). Sludge-fertilized lettuce may elevate hepatic microsomal oxidase activity in mice (Chaney et al., 1978a) while silage from sludge-grown corn can cause ultrastructural changes in the livers of sheep (Heffron et al., 1980). Otherwise, feeding sludge-fertilized grains and leaf crops to guinea pigs, rats, mice, swine, and pheasants has resulted in accumulation of nonessential trace elements but has failed to elicit any toxic responses (Furr et al., 1976b; Miller and Boswell, 1976; Chaney et al., 1978a,b; Hansen et al., 1976; Hinesly et al., 1976). It therefore seemed desirable to conduct a worst-case investigation short of the direct feeding of sewage sludge.

Such a study was approached by winter foraging weanling gilts for two seasons on experimental corn plots which had received heavy applications of industrial/residential sewage sludge for 8 years. A single season was not detrimental to the swine; however, during the second season which included breeding and gestation on the plots, those sows on moderately amended soils outperformed the controls while those on very heavily amended soils were adversely affected (Hansen and Hinesly, 1979).

The question of ill effects to animals through exposure to sludge-fertilized produce, though, is not as critical as the introduction of potentially toxic chemicals into the food web. Heavy metals, especially Cd, have been investigated, and Cd content is the primary factor considered in determining the suitability of sludges for use on agricultural soils (*Fed. Regist.*, 1979). Some Russian workers have been evaluating the safety of sludges containing high concentrations of solvents and other organic chemicals (Karyukhina et al., 1971; Khramova and Kutepov, 1974; Novoderzhkina et al., 1974), while Americans have been looking primarily at polychlorinated biphenyl (PCB) and chlorinated pesticide content of sludges (Furr et al., 1976a; Lawrence and Tosine, 1977; Erickson and Pellizzari, 1979).

PCB's are mixtures of randomly chlorinated biphenyl, there being 210 possible analogues with 0-10 chlorines per

biphenyl. In spite of the interest in and importance of PCB residues, nondescriptive estimation procedures based on selected Aroclor peaks are generally used for quantitation by researchers as well as regulatory agencies. Certain structures predominate, but the composition is not adequately predictable for accurate quantitation by this procedure. Erickson and Pellizzari (1979) have reported the distribution of chlorobiphenyls in a municipal sludge by degree of chlorination and this is certainly more informative. In examining the PCB content of the treated soils, we found the estimation by measuring selected Aroclor peaks unsatisfactory; therefore, in measuring the residues in swine fat we opted for the more specific quantitation of the individual chlorobiphenyls which could be identified.

### MATERIALS AND METHODS

**Animal Treatment.** The exposure area consisted of 16 (four application rates  $\times$  four replicates) 6.1  $\times$  12.2 m corn plots which had received fractions (0,  $\frac{1}{4}$ ,  $\frac{1}{2}$ , or 1) of the maximum amount of liquid digested Chicago sewage sludge which could be applied during the previous eight growing seasons and during the intervening season. The average annual furrow application rate for liquid digested sludge on the maximum-treated plots was 21.4 cm, with a range of 12.7 cm (1972 and 1975) to 38.1 cm (1971), depending on weather conditions. These application rates resulted in a 9-year total of 0, 126, 252, and 504 metric tons (t) of dry solids per hectare for zero, one-fourth, half, and maximum treatments, respectively (Hinesly and Hansen, 1979). Stalks, leaves, and cobs were returned to the respective plots each year after assessing production and removing grain and analytical samples.

Sixteen purebred Berkshire gilts were obtained from Thurow Farms, Yorkville, IL, and assigned to the experimental plots in a random fashion, except that no siblings were permitted within the same dosage group. The gilts were introduced in Nov 1975, removed in March 1976, and bred in early summer. They farrowed in October and were returned to the plots as sows in December where they were bred in January. They were removed in March for the second farrowing. Animals were provided with individual shelters, feeders, and waterers and straw bedding during January and February. They were fed standard hog rations appropriate for the stage of growth, gestation, and nursing.

**Elemental Analysis.** Soils were sampled and analyzed by flame atomic absorption spectrometry (Hinesly et al., 1977). Swine tissue samples were dehydrated and dry ashed at 450 °C, solubilized with nitric acid, and then analyzed by atomic absorption. Cadmium binding protein was analyzed in hepatic, renal, and mucosa supernatants

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Table I. Composition of Soils Amended for Eight Years with Sewage Sludge<sup>a</sup>

analyte	% composition for plot treatment <sup>b</sup>					LSD <sup>c</sup>	analyte	composition, ppm, for plot treatment					LSD
	0 t/ha	126 t/ha	252 t/ha	504 t/ha				0 t/ha	126 t/ha	252 t/ha	504 t/ha		
ash	95.5	95.8	94.9	93.8	1.3**	Zn	82	159	265	434	127**		
C	1.70	1.77	2.28	2.83	0.71**	Cr	37.5	76.9	129.4	228.2	79**		
Fe	1.93	1.85	1.95	2.14	NS	Pb	37.7	50.3	82.5	130.8	41**		
K	1.83	1.88	1.83	1.77	NS	Cu	18	41	72	122	33**		
N	0.143	0.139	0.184	0.234	0.062**	Ni	16.6	22.5	27.6	37.0	8.0**		
Na	0.820	0.824	0.809	0.800	NS	Cd	0.68	4.59	9.90	19.43	6.42**		
Ca	0.643	0.675	0.729	0.800	NS	pH	7.0	7.0	6.8	6.4	0.4*		
Mg	0.432	0.425	0.442	0.423	NS								
Mn	0.162	0.129	0.141	0.154	NS								
P	0.078	0.108	0.181	0.294	0.095**								

<sup>a</sup> Means of four replicate samples. <sup>b</sup> Cumulative dry solids added in 9 years. <sup>c</sup> Least significant difference; (\*) significant difference at 5% level; (\*\*) significant at 1% level; NS = not significant.

after separation on Sephadex G-75 (Washko and Cousins, 1976). Column fractions were acidified and analyzed for Cd by graphite furnace atomic absorption.

**Organochlorine Analysis.** Ethyl acetate extracts of soil samples were analyzed for PCB by the Illinois Natural History Survey Pesticide Laboratory after Florisil cleanup with hexane elution. Quantitative estimates were based on three and four representative peaks in Aroclor 1242 and 1254, respectively (Figure 1), and recovery was greater than 97%.

For pesticide analysis in animals, 5.0-g samples of back fat and bone marrow were hexane-extracted and left at -15 °C overnight to precipitate fat. After vacuum filtration with chilled apparatus, the extract was acetonitrile partitioned, concentrated, and placed on a Florisil column. Elution with ethyl ether in hexane resulted in >91% recovery from seeded fat for all pesticides reported. After GLC analysis, the samples were all pooled (equivalent to about 32 × 5.0 or 160 g of fat) and concentrated for GC-MS screening. Mass spectral analysis was conducted by the Environmental Research Laboratory, University of Illinois, to confirm the identity of specific peaks.

Separate 5.0-g fat samples were analyzed for PCB at the State Institute for Quality Control of Agricultural Products (RIKILT), Wageningen, The Netherlands. The fat was saponified, cleaned by alumina column chromatography, and analyzed by splitless glass capillary gas chromatography (Tuinstra et al., 1980). Individual chlorobiphenyls were quantitated with pure standards after matching retention times with pentachloroaniline (PCA) and mirex internal standards. Molecular weights of extract peaks were confirmed by mass spectrometry to conform to the molecular weight of the chlorobiphenyl standards. Many pesticides are destroyed by the saponification procedure. DDT is converted to DDE, but DDE, hexachlorobenzene (HCB), endrin, and dieldrin are not affected; however, endrin and dieldrin are withheld on the alumina column, further reducing interference.

## RESULTS AND DISCUSSION

**Soil Residues.** The soils on the experimental plots have been thoroughly characterized since sludge applications were initiated in 1968 [e.g., Varanka et al. (1976) and Hinesly and Hansen (1979)]. Table I presents analytical data from the soils for the spring of 1976, after the gilts were removed. In addition to graded increases in nutrient and nonnutrient elements, organic matter increased and pH decreased with increasing sludge applications.

PCB concentrations in the soils must be considered only estimates (Table II). Values for Aroclor 1242 will be exaggerated because several peaks were absent or greatly reduced, possibly due to microbial metabolism of the lower chlorinated components (Furukawa and Matsumura,

Table II. Estimated PCB Residues in Soils Amended for Eight Years with Sewage Sludge

treatment <sup>a</sup>	n	concn, mg/kg dry wt, ± SD		
		Aroclor 1242 <sup>b</sup>	Aroclor 1254 <sup>c</sup>	total
0	3	1.47 ± 0.27	0.15 ± 0.03	1.62 ± 0.29
126	4	1.72 ± 0.26	0.16 ± 0.02	1.88 ± 0.27
252	3	1.83 ± 0.50	0.30 ± 0.11	2.13 ± 0.51
504	4	2.19 ± 0.12	0.62 ± 0.15	2.81 ± 0.25

<sup>a</sup> Cumulative dry solids added (metric tons per hectare).

<sup>b</sup> Estimated from three peaks. <sup>c</sup> Estimated from four peaks.

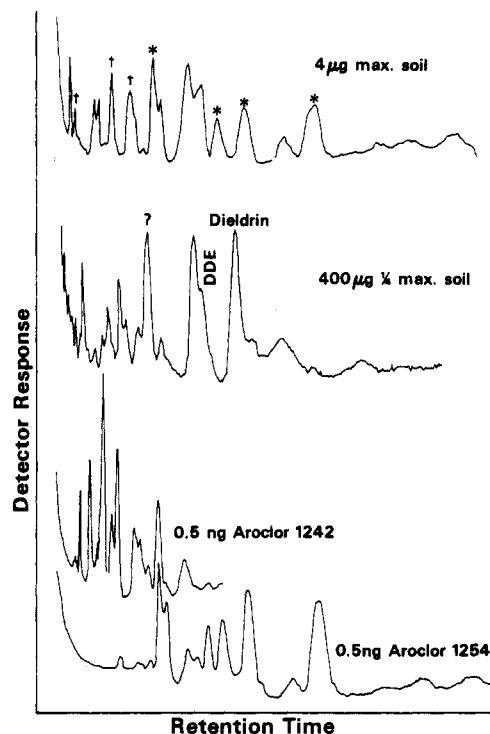
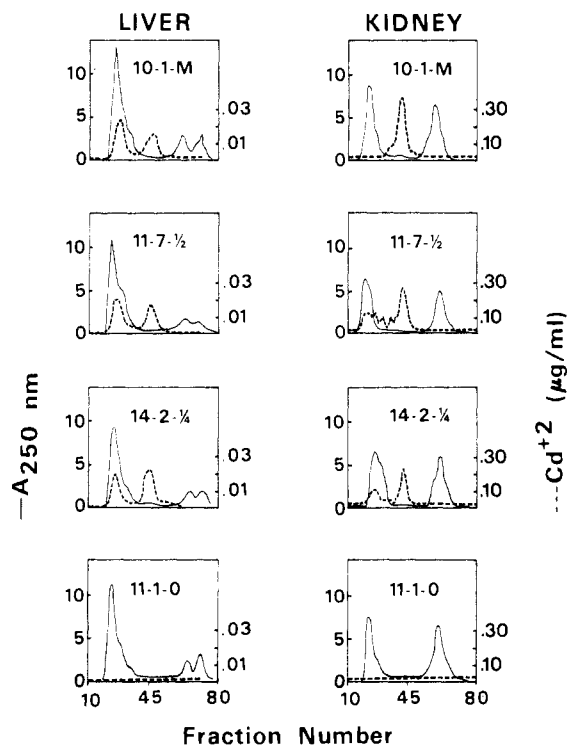


Figure 1. Gas-liquid chromatograms of Aroclors and representative soil extracts appropriately diluted. Peaks labeled with crosses were used to quantitate Aroclor 1242 while asterisks were used for Aroclor 1254.

1976); however, many of the lower chlorinated peaks were still present (Figure 1), indicating that Aroclor 1242 did contribute significantly to the residues. In the New Bedford, MA, sludge examined by Erickson and Pellizzari (1979), ~90% of the residue was, indeed, determined to be di-, tri-, and tetrachlorobiphenyls; on the other hand, Lawrence and Tosine (1977) estimated that Canadian sludges were mainly contaminated by Aroclors 1254 and 1260.



**Figure 2.** Protein and cadmium profiles of renal and hepatic supernatants from representative sows of each dose group. (—)  $A_{250\text{nm}}$  is maximal for CdBP; (---) Cd ( $\mu\text{g}/\text{mL}$ ) scale is different for hepatic vs. renal fractions.

**Soil Consumption.** While on the plots, the animals readily rooted and wallowed in the sludge-laden soils and consumed the plant parts returned to the fields. Rooting and foraging activity was considerably lower during January and February, particularly during the rather severe winter of 1977. Fecal excretion of certain elements (Table III) was used to confirm that the animals were actually ingesting the sludge-laden soils. Fecal concentrations of heavy metals (particularly Cr, Ni, and Pb, since they are poorly absorbed) reflect the pattern of greater soil ingestion in mid-March than in mid-February and rapid return to ambient levels when sows were removed from the plots and placed on wooden floors. Fecal concentrations reflected soil concentrations and, thus, sludge application rates except for a disproportionate increase in fecal residues between half-maximum (252 t/ha) and maximum (504 t/ha). Soil and fecal Mn was not influenced by sludge application rates, but fecal Mn declined after soil access was denied.

**Accumulation of Metals in Tissues.** Most of the Zn excess in the sludge-amended soils was apparently absorbed by the sows (Table III) but did not accumulate in tissues (Table IV). Ca, Cr, and Mg were also analyzed but showed no differences between treatment groups. A possible nonlinear relationship between soil Fe, tissue Fe, and biological consequences has been discussed previously (Hansen and Hinesly, 1979). The only element which showed dose-related accumulation was Cd in the kidney, liver, spleen (Table IV), and pancreas (data not included). The mean Cd concentration in maximum-exposed sows was slightly attenuated due to inclusion of a replacement sow which spent only one season on the plots (see footnote c of Table IV).

**Cadmium Binding Protein.** The elution profiles of soluble proteins from renal supernatants (Figure 2) were very similar to those described previously for swine (Cousins et al., 1973). Increases in cadmium binding protein (CdBP) were not seen in the Cousins study at 50

**Table III.** Mean Concentrations of Selected Elements in the Feces from Sows during and after Overwintering on Sludge-Amended Soils

treatment <sup>a</sup>	n <sup>b</sup>	concn, mg/kg fresh wt						
		Cd	Cr	Cu	Mn	Ni	Pb	Zn
February (on Plots)								
0	2	0.59	4.4	22	130	3.6	5.0	225
126	3	0.49	5.3	23	69	3.1	3.1	235
252	2	0.85	8.2	25	96	3.6	5.8	228
504	4	7.19	67.0	55	176	12.1	35.4	286
March (on Plots)								
0	3	1.16	4.3	19	250	4.3	7.9	209
126	4	1.23	— <sup>c</sup>	—	—	—	—	—
252	4	2.33	19.7	36	176	6.0	11.4	315
504	1	9.01	82.2	64	253	13.0	41.7	336
March (4-6 Days after Removal from Plots)								
0	4	0.20	1.2	16	32	1.1	1.0	171
126	4	0.20	1.6	16	45	1.7	1.2	174
252	4	0.20	1.4	15	30	1.2	2.0	153
504	4	0.45	1.7	16	35	1.4	1.0	152

<sup>a</sup> Cumulative dry solids (metric tons per hectare) in 10 years. <sup>b</sup> Number of discrete, fresh samples available. <sup>c</sup> This series of samples was lost.

**Table IV.** Mean ( $n = 4$ ) Concentrations of Selected Elements in Tissues from Sows Overwintered for Two Seasons on Sludge-Amended Soils

treatment <sup>a</sup>	concn, mg/kg fresh wt						
	Cd	Cu	Fe	Pb	Mn	Ni	Zn
Kidney							
0	0.35	5.3	69	0.25	1.03	<0.18	27
126	1.90	3.7	110	0.26	1.01	<0.18	28
252	3.69	5.5	78	0.28	0.96	<0.18	30
504 <sup>c</sup>	4.71	6.4	66	0.40	0.90	<0.18	31
LSD	2.88 <sup>b</sup>						
Liver							
0	0.03	6.2	150	0.26	1.87	0.19	57
126	0.33	13.2	211	<0.18	1.83	<0.18	58
252	0.42	3.5	202	0.32	1.96	<0.18	64
504	0.83	5.4	158	<0.18	1.57	<0.18	52
LSD	0.52						
Muscle (Ham)							
0	<0.02	0.7	14	<0.18	0.09	<0.18	36
126	<0.02	0.7	16	0.25	0.07	<0.18	36
252	0.02	0.6	15	0.43	0.11	<0.18	37
504	0.03	0.6	15	0.28	0.12	<0.18	34
NS <sup>d</sup>							
Heart Muscle							
0	<0.02	3.4	30	0.22	0.22	<0.18	17
126	0.03	3.6	23	<0.18	0.23	<0.18	16
252	0.02	4.0	42	0.36	0.24	<0.18	17
504	0.03	2.6	29	<0.18	0.22	<0.18	16
NS							
Spleen							
0	<0.02	0.7	400	0.24	0.21	<0.18	27
126	<0.02	0.7	503	<0.18	0.54	<0.18	28
252	0.04	0.8	595	0.37	0.28	0.19	27
504	0.10	1.5	507	0.23	0.23	<0.18	26
LSD	0.05						

<sup>a</sup> Cumulative dry solids (metric tons per hectare) in 10 years. <sup>b</sup> Least significant difference at  $P < 5\%$ . <sup>c</sup> Includes data from replacement sow having spent a single season on the plots. Mean Cd for  $n = 3$  is as follows: kidney =  $5.52 \pm 0.37$ ; liver =  $0.96 \pm 0.21$ . <sup>d</sup> No significant differences.

mg/kg in the diet, although mean renal residues were 41.2 mg/kg fresh weight (equivalent to more than 160 mg/kg dry weight). Measuring absorbance at 250 nm, we were also unable to consistently detect CdBP in our samples, but the Cd concentrations were in the optimum analytical

Table V. Mean Concentrations of Chlorinated Hydrocarbons in Fatty Tissues from Sows Overwintered Two Seasons on Sewage Sludge Amended Plots

8-year sludge treatment	n	chlorinated hydrocarbon				n	$\mu\text{g}/\text{kg}$ fat basis <sup>a</sup> $\pm$ SD	
		concn, $\mu\text{g}/\text{kg}$ fresh wt, $\pm$ SD					DDE	PCB
		lindane	heptachlor epoxide	dieldrin	<i>p,p'</i> -DDE			
control								
back fat	4	3 $\pm$ 2	7 $\pm$ 9	11 $\pm$ 8	12 $\pm$ 3	4	27 $\pm$ 5	36 $\pm$ 9
marrow <sup>b</sup>	4	5 $\pm$ 2	4 $\pm$ 3	6 $\pm$ 4	10 $\pm$ 5			
126 t/ha								
back fat	4	2 $\pm$ 1	2 $\pm$ 2	14 $\pm$ 10	21 $\pm$ 15	4	41 $\pm$ 18	106 $\pm$ 64
marrow	3	3 $\pm$ 3	8 $\pm$ 6	11 $\pm$ 3	15 $\pm$ 3			
252 t/ha								
back fat	3	<1	5 $\pm$ 8	25 $\pm$ 35	30 $\pm$ 16	4	51 $\pm$ 17	191 $\pm$ 97
marrow	4	5 $\pm$ 5	4 $\pm$ 4	8 $\pm$ 6	32 $\pm$ 22			
504 t/ha								
back fat	3	<1	<1	12 $\pm$ 12	101 $\pm$ 18	3	97 $\pm$ 6	389 $\pm$ 118
marrow	3	5 $\pm$ 4	<1	7 $\pm$ 5	58 $\pm$ 4			

<sup>a</sup> Determined by saponification and capillary column GLC of separate samples in the RIKILT Laboratory. DDE will be higher than in the previous column for reasons discussed in under Materials and Methods. Total PCB is the summation of 12 peaks (Table VI) determined individually. PCB values less than the detection limit of 5 ppb were calculated at 2.5 ppb.

<sup>b</sup> Bone marrow from femur which is ~66% fat.

Table VI. Concentration of Individual Chlorobiphenyls in the Back Fat of Sows Having Overwintered on Sludge-Amended Soils for Two Seasons

chlorobiphenyl structure	mean fat concn, $\mu\text{g}/\text{kg}$ on a fat basis, <sup>a</sup> $\pm$ SD			
	control (n = 4)	1/4 max (n = 4)	1/2 max (n = 4)	max (n = 3)
2,5,2',5'	<5	7.5 $\pm$ 1.9	10.5 $\pm$ 5.2	23 $\pm$ 10
2,3,6,2',5'	<5	5 $\pm$ 3.1	6.5 $\pm$ 5.0	13.7 $\pm$ 6.6
2,4,5,2',5'	<5	<5	5.2 $\pm$ 4.0	6.9 $\pm$ 3.8
2,3,5,6,2',5'	<5	<5	3.6 $\pm$ 2.2	3.6 $\pm$ 2.2
2,4,5,3',4'	<5	<5	6.8 $\pm$ 5.0	10.5 $\pm$ 7.1
2,4,5,2',4',5'	4.1 $\pm$ 3.2	23.8 $\pm$ 13.8	31.2 $\pm$ 16.7	89.3 $\pm$ 23.4
2,3,4,2',4',5'	5.5 $\pm$ 4.0	27.0 $\pm$ 17.6	36.8 $\pm$ 19.5	110.3 $\pm$ 34.9
2,3,5,6,2',4',5'	<5	6.4 $\pm$ 4.0	8.4 $\pm$ 5.2	23.3 $\pm$ 6.6
2,3,4,2',3',4'	<5	7.5 $\pm$ 8.4	6.6 $\pm$ 4.0	17.7 $\pm$ 6.0
2,3,4,5,2',4',5'	<5	13.8 $\pm$ 8.9	21.0 $\pm$ 11.4	58.3 $\pm$ 18.8
2,3,4,5,2',3',4'	<5	4.1 $\pm$ 3.2	6.6 $\pm$ 3.7	15.0 $\pm$ 9.8
2,3,4,5,2',3',5',6'	<5	3.4 $\pm$ 1.8	5.1 $\pm$ 2.2	11.0 $\pm$ 4.5

<sup>a</sup> Concentrations below the limit of detection (5 ppb) were calculated as 2.5 ppb.

range for graphite furnace atomic absorption spectroscopy; therefore, the presence of minute amounts of CdBP were confirmed in the livers and kidneys of representative treated sows (Figure 2).

Sow 14-2 accumulated the highest liver Cd residue of the four sows represented, and the slight increase in absorbance at 250 nm around fraction 40 indicates the presence of CdBP; this is verified by the greater proportion of Cd in these fractions than in the earlier fractions. A more striking Cd shift was seen in the kidney fractions from sow 10-1 where a slight increase in protein absorbing at 250 nm is apparently adequate to sequester nearly all of the intracellular Cd. The 1000-fold increase in sensitivity for Cd failed to reveal any CdBP in the liver or kidneys from the control sow (Cd < 0.1 ng/mL in liver fractions and < 20 ng/mL in the kidney fractions). As might be expected, the lag between removal of the sows from the experimental soils and termination at weaning was too great to preserve any CdBP in the intestinal mucosa of any sows.

**Pesticide Residues.** Table V presents a summary of chlorinated hydrocarbon residues in the back fat of the sows. Aldrin and dieldrin had been used for corn rootworm control in the early years of the study so the dieldrin residues are equally distributed among all groups and did not originate from the sewage sludge. A single fat sample from the 252 t/ha group contained 66 ppb of dieldrin and may have resulted from contamination since the bone

marrow sample contained only 16 ppb of dieldrin. Likewise, hexachlorobenzene (HCB), lindane, and heptachlor epoxide residues could not be attributed to the sewage sludge. HCB data are not presented because four sows farrowed and nursed litters in crates previously occupied by sows receiving 20 ppm of dietary HCB. These four sows accumulated 40–220 ppb of HCB in their fat; however, this should not have contributed to any observed effects since it is only 0.25% of the residue accumulated by the HCB-fed sows which exhibited only mild signs of toxicosis (Hansen et al., 1979). HCB residues were between 5 and 15 ppb in sows not farrowed in former HCB pens.

Resolution of *p,p'*-DDT and *p,p'*-DDD from interfering peaks was inconsistent and not reportable; nevertheless, it is apparent from both sets of samples that *p,p'*-DDE accumulation was proportional to sludge application (Table V). The samples processed in the RIKILT Laboratory would be expected to yield slightly higher numbers because DDT was converted to DDE and dissectable fat from sows may contain only ~80% pure fat (Hansen et al., 1979).

Other organics detected by GLC-mass spectrometry of concentrated fat extracts, but not quantitated, included hexachlorocyclohexane (BHC) isomers in addition to lindane, dibutyl and dioctyl phthalate, and endrin.

**Polychlorinated Biphenyls.** The PCB components were identified by cochromatography with pure chlorobiphenyl standards (Figure 3). Such a procedure underestimates total PCB since not all peaks were identified,

Table VII. Chlorobiphenyls Included in the Standard Mixture but Which Were Not Present above the Detection Limit

structure	detection limit, mg/kg
2-monochlorobiphenyl	0.05
4-monochlorobiphenyl	0.05
2,2'-dichlorobiphenyl	0.05
4,4'-dichlorobiphenyl	0.025
2,4-dichlorobiphenyl	0.005
3,4,2'-trichlorobiphenyl	0.005
2,4,2',5'-tetrachlorobiphenyl	0.005
2,3,2',5'-tetrachlorobiphenyl	0.005
2,5,2',4'-tetrachlorobiphenyl	0.005
2,4,5,2',3'-pentachlorobiphenyl	0.005
2,3,4,2',5'-pentachlorobiphenyl	0.005
2,3,6,2',3'-pentachlorobiphenyl	0.005
2,3,4,5,3',4'-hexachlorobiphenyl	0.005
2,3,6,2',3',6'-hexachlorobiphenyl	0.005
2,3,4,5,2',5'-hexachlorobiphenyl	0.005
2,3,4,5,6,2',5'-heptachlorobiphenyl	0.005
2,3,5,6,2',3',5',6'-octachlorobiphenyl	0.005
2,3,4,5,2',3',4',5'-octachlorobiphenyl	0.005
2,3,4,5,6,2',3',4',5'-nonachlorobiphenyl	0.005

but the treatment-related accumulation is readily apparent (Figure 3 and Table VI). More than half of the residue can be accounted for by 3 chlorobiphenyls (Table VI), and 19 chlorobiphenyls were found to be absent or below the limit of detection (Table VII). At these low concentrations many of the Aroclor 1242 components are metabolized (Hansen et al., 1977; Hansen, 1979); however, 2,5,2',5'-tetrachlorobiphenyl is the predominant component of Aroclor 1242, and its accumulation in the pig fat cannot be attributed to Aroclor 1254 (Webb and McCall, 1972; Hirwe et al., 1974). The high concentrations of 2,4,5,2',4',5'- and 2,3,4,2',4',5'-hexachlorobiphenyls in the fat are indicative of Aroclor 1254 or 1260 contamination, but the accompanying low concentration of 2,4,5,3',4'-pentachlorobiphenyl indicates that this component which normally persists in swine fat (Hansen and Welborn, 1977; Hansen, 1979) and is highly toxic (Ax and Hansen, 1975) was selectively degraded during the sludge digestion process or later in the soils. Alternatively, Aroclor 1254 may not have been involved since this component is present in low amounts in Aroclors 1242 and 1260 (Ballschmitter and Zell, 1980).

PCB estimation as specific Aroclors based on a few selected peaks is not applicable to PCB mixtures subjected to even simple biological and physical selection processes (Cook, 1972; Hansen et al., 1977; Hansen, 1979; Tuinstra and Traag, 1979; Ballschmitter and Zell, 1980; Sparling and Safe, 1980; Tuinstra et al., 1980, 1981; Zell and Ballschmitter, 1980). This method of estimation (not quantitation) is useful in detecting gross contamination when other interfering analytes are minimal and when PCB contents differ by large factors. In this investigation, the estimate method predicted 1.7-fold differences in PCB content between control and maximum treated soils. In reality, there was at least a 10.8-fold difference between total PCB content in fats (Table V); moreover, this difference probably would have been closer to the 20-fold factor seen for 2,4,5,2',4',5'- and 2,3,4,2',4',5'-hexachlorobiphenyls if the control fat residues could have been more accurately quantitated (Table VI).

The estimation procedure, while useful for indicating possible sources and severe differences, is unacceptable for determining the PCB content of sewage sludges and sludge-derived residues in the food web. This is because of the nonpoint source nature of the residues and because

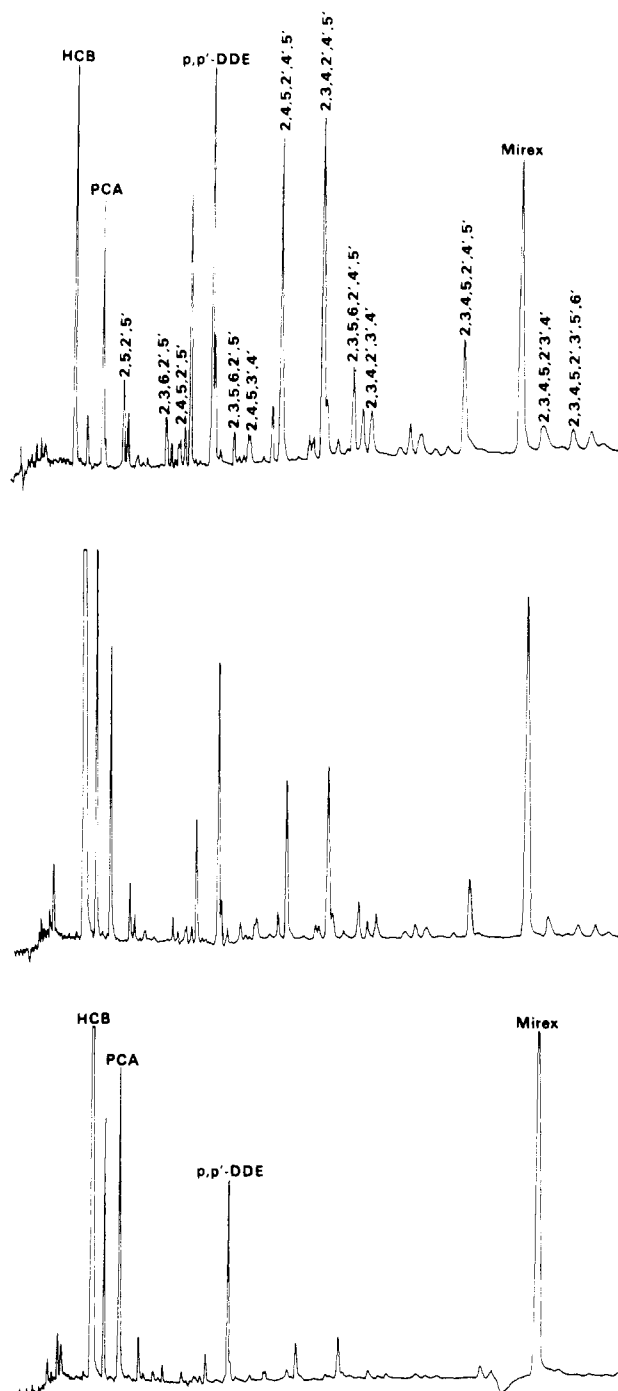


Figure 3. Capillary column gas chromatograms of the equivalent of 1 mg of back fat from control sow 9-2 (lower), one-fourth maximum sow 14-2 (middle), and maximum sow 14-1 (top). Mirex and PCA were added as internal standards.

of the complex physical, chemical, and biological processes involved in the treatment of sewage. The trend to ignore the historical realization that environmental PCB residues cannot be equated to commercial mixtures (Cook, 1972) is quite disturbing—the residues reported are unavoidably in error, but results can be reverently applied to ignore or amplify differences that cannot be accurately detected by these methods. It would be better to use perchlorination procedures and at least report accurate concentrations for total chlorobiphenyls than to use the estimation procedure. It would be much better to establish a complete series of standards and to precisely characterize the PCB residues which occur in various stages of the food web as a result of the land disposal of sewage sludges.

Table VIII. Cadmium and PCB Residues in Sows As Related to Nursing Activity, Soil Ingestion, and Sibling Relationships

animal (time) <sup>a</sup>	piglets <sup>b</sup> weaned	fecal Cr/soil Cr <sup>c</sup>	Cd, mg/kg dry wt		PCB, mg/kg	
			renal	hepatic	soil <sup>d</sup>	fat <sup>e</sup>
control						
11-1 (62)	8	0.07	0.24	0.03	0.15	0.032
10-4 (118)	7		0.32	0.02	0.18	0.034
12-4 (61)	1	0.08	0.38	0.05		0.030
9-2 (65)	0	0.20	0.45	0.04	0.12	0.050
126 t/ha						
10-6 (114)	5	0.08	0.28	0.08	0.18	0.068
12-3 (71)	5	0.08	2.24	0.35	0.15	0.066
8-2 (93)	8	0.09	2.46	0.36	0.15	0.092
14-2 (93)	0		2.61	0.54	0.16	0.200
252 t/ha						
12-7 (69)	3	0.06	2.38	0.23		0.063
8-6 (69)	3	0.29	2.88	0.45	0.37	0.170
11-7 (65)	5	0.08	4.05	0.33	0.17	0.278
14-8 (61)	7	0.16	5.47	0.68	0.37	0.253
504 t/ha						
12-2 (62)	3	0.33	5.18	0.84	0.56	0.279
14-1 (98)	8	0.25	5.46	0.83	0.47	0.514
13-1 (71)	1	0.36	5.91	1.20	0.63	0.373
10-1 <sup>f</sup> (89)	5	0.22	2.29	0.45	0.82	0.126

<sup>a</sup> Sow number followed by days (in parentheses) between removal from plots and termination. <sup>b</sup> Number of piglets from the second litter nursed for the full 5 weeks. <sup>c</sup> Fecal chromium concentration within 1 day of removal from plots divided by chromium concentration in the soil should provide a reasonable estimate of soil ingested. <sup>d</sup> Estimate based on four peaks from Aroclor 1254. Aroclor 1242 predominated in all soils, however, but made only a small contribution to the residues in fat. <sup>e</sup> Summation of 12 chlorobiphenyls quantitated individually. <sup>f</sup> Replacement sow that spent only one season on the experimental plot.

**Sibling Comparisons.** If various relationships are considered individually, it can be seen that no single factor—not even sludge application rate—can account for the residues of Cd or PCB's accumulated in individual sows (Table VIII). The animal designations include an initial number (8–14 in this case) which indicates the mother, followed by a second number indicating the sequence of birth. Thus, sow 11-1 was the first in a litter while sow 11-7 was a sister born six piglets later. When sibling comparisons between treatment groups are possible, litters 10 and 12 seem to be more refractory to residue accumulation than litter 14 in spite of soil ingestion and soil PCB (12-2 vs. 14-1). A greater number of pigs weaned should reduce the PCB load since parturition and nursing would provide added routes of elimination, but compare sows 12-7 and 14-8 and, again, 12-2 vs. 14-1. Generally, renal Cd concentrations parallel PCB residues in the fat, but this cannot be related to estimates of soil ingestion (8-6 vs. 11-7 vs. 14-8). Perhaps genetics may play a role in food animal residues as it does in grain residues (Hinesly et al., 1978) and should be exploited.

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