

Plant Contamination by PCBs from Amended Soils

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ABSTRACT

Plants accumulated PCBs (Aroclors) from soil amended with lake sediment contaminated with Aroclors 1248, 1254, and 1260. Uptake into different parts of vegetable plants was investigated in the field by growing beets (*Beta vulgaris* L.), turnips (*Brassica rapa* L.) and beans (*Phaseolus vulgaris* L.). In beets and turnips, leaves accumulated larger concentrations of PCBs than the roots. In beans, leaves and pods contained higher concentrations than the stems, while only low concentrations were detected in the seeds. Bioaccumulation of Aroclors by plants was in the following order: Aroclor 1248 > 1254 > 1260. Relative to their concentration in the soil, the lower chlorinated PCB isomers which are more soluble in water and more volatile were more abundant in plants than the higher chlorinated isomers.

PCBs (polychlorinated biphenyls), a group of toxic and persistent organic compounds, have been extensively used in a number of industrial products. Although no longer manufactured in this country, their prior manufacture, use and disposal have resulted in their entry into the environment, particularly in sediments of streams and rivers. Fish caught from contaminated water contain PCBs in excess of the 5 mg/kg tolerance limit established by the U.S. Federal Food and Drug Administration (4). Tolerances for other food products vary from 0.3 mg/kg in eggs to 1.5 mg/kg (on a fat basis) in milk and manufactured dairy products (4). Presence of PCBs in river sediments continues to be a source for their entry into the food chain. A possible method to decontaminate rivers is to dredge and dispose the sediments on land. However, it is essential to know whether plants growing on contaminated land would take up the PCBs, and thus become another pathway for their entry into the food chain.

PCBs manufactured in the U.S. have been marketed under the trade name Aroclors (Monsanto Chemical Co.). Most common preparations of Aroclors are 1242, 1248, 1254, and 1260; the last two digits indicate the average percentage of chlorine in the PCB molecule. Each Aroclor preparation contains a number of chlorobiphenyl isomers.

Investigations of PCB uptake by plants are sparse and mostly limited to either one or more individual chlorinated biphenyl isomers or a single PCB preparation added to the growth medium. Iwata et al. (9), in studies of uptake of Aroclor 1254 by carrots, found that 97% of the

Aroclor taken up from the soil was retained by the carrot peels. These researchers also detected the Aroclor in the foliage and attributed its presence primarily to absorption of contaminated soil dust. Fries and Marrow (5) used individual isomers in their study and attributed their presence in the foliage of soybean plants to contamination from vapor sorption. Accumulation of air-borne PCBs by plant foliage has recently been demonstrated by Buckley (2). Experiments with ^{14}C -labelled Aroclor 1254 showed only small concentrations in soybean and fescue (20). Orchard grass and carrots grown in a soil contaminated with PBBs (polybrominated biphenyls) showed no or very minor uptake (11). Similarly, Chou et al. (3) found that when soybean and corn seedlings were grown in a nutrient medium containing ^{14}C -labelled PBB isomers for about a 1-week period, plant roots contained radioactive residues but no radioactivity was detected in the leaves. These authors also found PBB in concentrations ranging from about 38 to 535 $\mu\text{g/kg}$ in radish, carrot, and onion roots grown in two soils contaminated with 100 mg of PBB/kg. Above ground portions of the crops, however, were not analyzed in these experiments.

In contrast to these investigations, Babish et al. (1) found that cabbage grown on a 120-cm deep deposit of anaerobically digested sewage sludge contaminated with PCBs contained about 400 μg of PCBs/kg and the liver from guinea pigs fed on this cabbage showed higher PCB concentrations than the liver from guinea pigs fed on cabbage from the control plots. In a study by Strek et al. (16), beet and peanut tops accumulated as much as 815 and 473 μg of ^{14}C -labelled Aroclor 1254/kg, respectively, from a sandy soil to which 20 mg of Aroclor/kg was added. Uptake of ^{14}C -labelled di- and tri-chlorobiphenyl isomers by spruce seedlings over a 4-year growth period, as determined from total radioactive residue, was 401 $\mu\text{g/kg}$ and 231 $\mu\text{g/kg}$ in needles and stems, respectively (13). In a sand culture experiment, Suzuki et al. (17) detected 150 $\mu\text{g/kg}$ in soybean sprouts grown in sand treated with 100 mg of Aroclor 1242/kg. These authors inserted a 7-cm thick layer of uncontaminated sand through which the soybeans grew into the contaminated sand. As the analysis of the "barrier" sand showed no PCBs, the uptake by the bean sprouts was considered to have occurred by PCB translocation through the plant roots.

Thus PCB uptake by crop plants growing on a contaminated growth medium have been shown to occur either through sorption of the volatilized PCBs or through their translocation via root uptake. Quantitative determination of the uptake of different Aroclors from contaminated environmental sources containing mixtures of Aroclors, however, has not been attempted. In the present investigation, we determined uptake of different Aroclors, as well as individual chlorobiphenyl isomers present in the Aroclors, by plants grown in field soil amended with lake sediment containing Aroclors 1248, 1254 and 1260.

MATERIALS AND METHODS

Source of PCBs and soil amendment

Bottom sediment from Woods Pond, which is contaminated with PCBs and is obviously a source of PCBs in the Housatonic River system (6), was used as an amendment for soil on which vegetables were grown. The sediment contained 4.6 mg of Aroclor 1248, 8.0 mg of Aroclor 1254, and 44.5 mg of Aroclor 1260/kg on a dry-weight basis. Seven hundred and fifty-six liters of sediment (about 30% solids) were poured on the surface of a 2 m x 2 m soil plot isolated in the field by 2.5 cm x 15 cm wooden boards inserted into the ground. When the amended plot appeared air-dried, the surface 15 cm of soil was thoroughly mixed. A control plot was similarly constructed and treated with water instead of sediment.

Sampling and preparation of plant material

In May 1981, turnips (*Brassica rapa* L.) and red beets (*Beta vulgaris* L.) were planted in each plot. Both leaves and roots were harvested on October 15, 1981, washed with warm water while scrubbing with a brush to ensure removal of any adhering soil particles. All plant material was dried in a forced-air oven at 70°C. Tests showed no loss of PCBs by drying plant material at this temperature. The dried parts were then ground in a stainless steel Wiley mill to pass through a 20-mesh sieve. In May 1982, turnips and snap beans (*Phaseolus vulgaris* L.) were planted in the same plots without any further addition of sediment. Turnip leaves were harvested three times during the growing season, whereas beans were harvested only once.

Extraction and analyses of PCBs

Ten-g samples of dried, ground plant material were extracted with 150 ml of petroleum ether and 20 g of the amended soil were extracted with 150 ml 1:1 acetone:hexane mixture. The extracts were purified to remove interfering chlorinated compounds (18,19) and analyzed with a gas chromatograph (GC) equipped with a ^{63}Ni electron capture detector. A 180-cm glass column (2 mm i.d.) filled with 3% OV-1 on Chromosorb W (80/100 mesh) was used. Details of the purification of extracts, operating conditions of the chromatograph and the method for determination of different Aroclors from the resulting chromatograms are described in an earlier publication (15).

RESULTS AND DISCUSSION

Uptake of PCBs from amended soil

Figure 1 shows gas chromatograms of PCB extracts from soil amended with sediment containing PCBs and from leaves of bean plants grown on the amended soil and on control soil. Leaf extracts were concentrated to 2 ml, whereas the soil extract was diluted to 200 ml to obtain optimum peak heights. Peaks 1 through 20 correspond to different chlorobiphenyl isomer peaks present in individual preparations of Aroclors 1248, 1254, and 1260 (Table 1). The lower numbered peaks with lower retention times (RTs) correspond to the less chlorinated biphenyls and higher numbered peaks to more chlorinated biphenyls. The data in Table 1 also show that the peaks obtained from individual preparations of the three Aro-

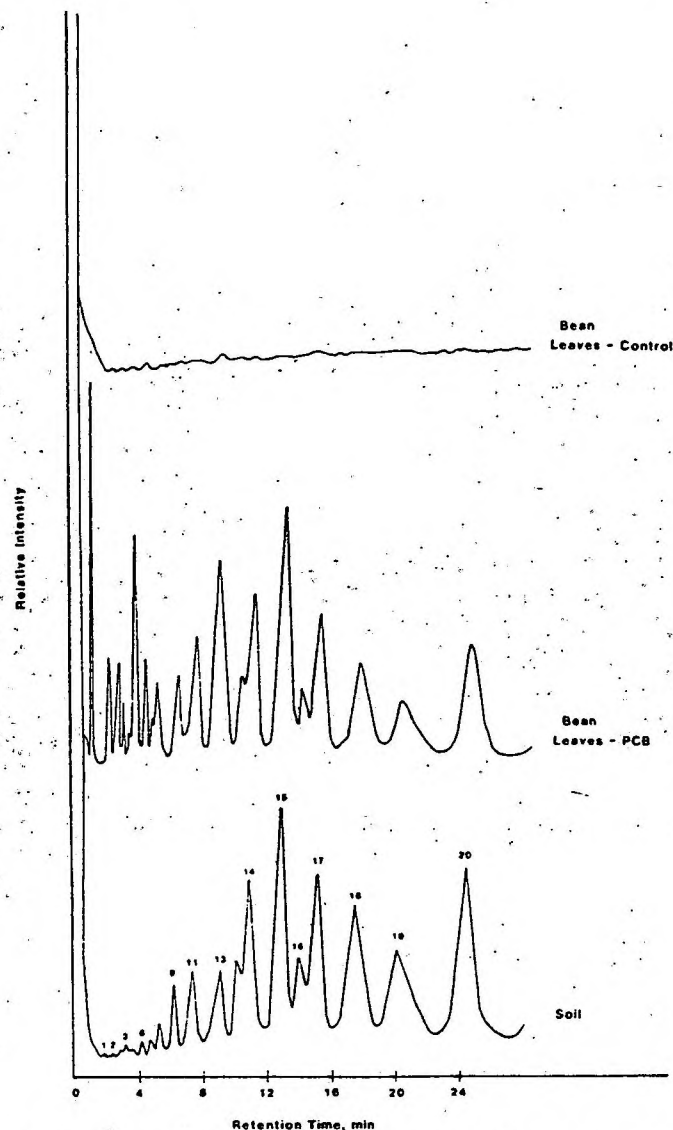


Figure 1. Gas chromatograms of extracts from field soil amended with lake sediment contaminated with PCBs and from leaves of bean plants grown on contaminated and control soils.

lors overlap, with peaks of lower chlorinated biphenyls more abundant in Aroclor 1248 and peaks of higher chlorinated biphenyls more abundant in Aroclor 1260. The peaks in the gas chromatograms (Fig. 1) clearly show that leaves of the bean plant grown in the amended soil contained PCBs that were present in the amended soil while the bean plants grown on the control soil contain no detectable PCBs. Heights of the different peaks, however, show that leaves contain relatively larger concentrations of the lower chlorinated biphenyls and smaller concentrations of the higher chlorinated biphenyls than the amended soil.

Bioaccumulation of different Aroclors from soil

Uptake of different chlorobiphenyls by plants in relation to their contents in the amended soil is illustrated in Fig. 2 by ratios of peak heights of individual chlorobiphenyls in extracts from the plants to those from the soil. The ratios appear to decrease with increasing retention times. Ratios of the low chlorinated biphenyls (the first 7 peaks), in leaves, to corresponding biphenyls in soil, are greater than 1, while ratios of the remaining biphenyls are less than 1 and decrease with in-

creasing chlorination. The relationship between the ratio of the GC peaks in leaves to peaks in soil and retention times of peaks (Fig. 2) show that under experimental conditions used, bioaccumulation by plants of chlorobiphenyls with retention times less than 12 min increases rapidly with decrease in chlorination. Bioaccumulation of the less chlorinated biphenyls corresponding to the first seven GC peaks was 20-60 times as high as the most chlorinated biphenyl corresponding to the last peak. Other parts of these plants and other plant species also showed uptake of larger concentrations of

TABLE 1. Heights of gas chromatographic peaks obtained from individual standard preparations of Aroclors 1248, 1254 and 1260^a.

Peak number	Height, mm		
	1248	1254	1260
1	4	-	-
2	18	-	-
3	10	-	-
4	42	-	-
5	16	-	-
6	25	14	-
7	23	7	-
8	21	2	-
9	41	23	8
10	16	2	3
11	9	21	9
12	6	12	4
13	8	19	6
14	6	20	22
15	4	15	29
16	-	3	13
17	-	12	20
18	-	3	16
19	-	2	12
20	-	2	19

^a2 µl of 0.2 µg/ml standard solution of each Aroclor were used.

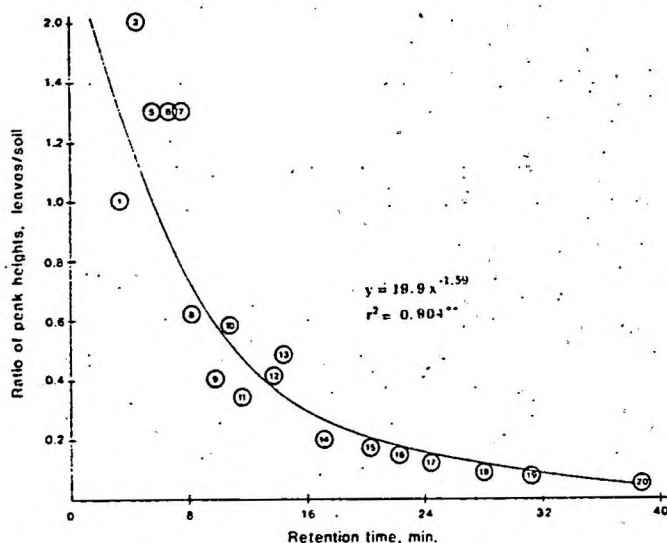


Figure 2. Ratios of GC peak heights from extracts of beet leaves grown on a soil amended with lake sediment contaminated with PCBs to the corresponding peaks from soil extracts. Numbers within the circles indicate peak number (see Table 1). Ratios for peaks 2 and 4 are not shown in figure because they lie beyond the selected graph scale.

the lower chlorinated biphenyls. These results are in agreement with observations of Iwata et al. (9) and Suzuki et al. (17) who used individual Aroclors and found that peaks with early retention times, corresponding to less chlorinated biphenyls, were higher in carrots and soybeans.

Estimates of amounts of different Aroclors, 1248, 1254, and 1260, obtained from analyses of peak heights (15) in extracts from different plant materials harvested in 1981 are given in Table 2. Comparison of concentrations of different Aroclors in roots and leaves of both beets and turnips suggest that leaves accumulate greater concentrations of each of the three Aroclors; for example, beet roots contain 15, 16, and 35 µg/kg, respectively, of Aroclors 1248, 1254, and 1260 while beet leaves contain 22, 94, and 52 µg/kg of the three Aroclors. Total concentrations of the three Aroclors in beet roots and leaves and in turnips roots and leaves were 66 and 168 and 66 and 99 µg/kg, respectively. These results are in contrast to those of Iwata and Gunther (10) who found much higher concentrations of Aroclor 1254 in the carrot roots than in the foliage of carrots grown on a soil to which 100 µg/kg Aroclor 1254 was added.

Ratios of the concentrations of different Aroclors in the plants grown in 1981 to their concentrations in the soil, the bioaccumulation factor (BF), show that bioaccumulation by plants was Aroclor 1248 > 1254 > 1260 (Table 2). The relative bioaccumulation factor (RBF), i.e. the BF factor of Aroclor 1248 or 1254 compared to Aroclor 1260, shows that bioaccumulation of Aroclor 1248 was 94 times more than 1260 by beet roots and 188 times more by turnip roots. Bioaccumulation of Aroclor 1254 was 4 times more than 1260 by the two root systems. The RBF for Aroclor 1248 was 69 for beet leaves and 200 for turnip leaves and for Aroclor 1254 it was 12 for beet leaves and 11 for turnip leaves. These trends would be expected whether PCBs are sorbed by the leaves as vapors or translocated through the root system via passive transport in the water stream because both the vapor pressure and the solubility of the Aroclors are in the order 1248 > 1254 > 1260 (8).

Concentrations of PCBs in various parts of the vegetables grown in the amended soil during the second year are given in Table 3. A comparison of the PCB concentrations in turnips grown in the amended soil in the two years (Table 2 vs. Table 3) shows a decrease in Aroclor 1248 uptake relative to Aroclors 1254 and 1260 in the second year. This decrease is likely due to a large reduction in the amount of Aroclor 1248 in the soil after the first year; indeed, only traces of Aroclor 1248 were detected in soil during the second year of cropping, and hence, no certain measurable concentration of 1248 in the second year is given here. Iwata et al. (9) also noted the lower GC peaks corresponding to less chlorinated biphenyls in the Aroclor 1254 applied to soil disappeared faster than higher peaks; a 20% decrease occurred in peak number 1 in 10 months, whereas peak 10 remained essentially the same during this period. Greater losses of the less chlorinated biphenyls can occur either by volatilization or by degradation. Our experiments (7) on microbial degradation of PCBs in soil cultures show that the less chlorinated

biphenyls degraded more readily in soils. Also, the degree of degradation was different in different soils and varied with cropping history of the soil. For example, 90% of Aroclor 1248 in sediment added to a fine sandy loam soil in corn was degraded in a 2-week period, while only 20% was degraded in a similar soil in a pine forest. The losses of PCBs due to volatilization would be minimal in culture experiments because the reaction was carried out in flasks stoppered with cotton plugs that prevented direct exposure of the reaction mixture to the atmosphere.

Although the mechanism of PCB degradation is not clear, ease of degradation of the less chlorinated Aroclors may be related to their higher solubility in water.

Data in Table 3 show that turnip leaves harvested on three different dates contained similar concentrations of the Aroclors, except that Aroclors 1254 and 1260 were somewhat higher in the last harvest. Greater concentrations in unpeeled than peeled turnips are in accord with results with unpeeled and peeled carrots (9). High concentrations in peels of crop roots likely result from diffusion and dissolution of PCBs in organic constituents of peels which are in close proximity to contaminated soil particles. Presence of PCBs in leaves, stems, pods and small amounts in seeds may be attributed, at least in part, to their uptake and translocation into various plant parts. These results and uptake of PCBs by soybean sprouts grown in sand culture (17) and their detection in rice grains (14), demonstrate that PCBs can be taken up by plant roots and translocated to above ground parts of plants. The mechanism by which these non-polar, neutral large organic molecules are taken up and translocated into the plant is not clear at present, but must be similar to uptake of organochlorine pesticide residues in soils (12).

Results of this investigation thus show that plants grown in soil contaminated with PCBs can absorb these compounds. However, the amounts taken up by the plant are relatively small compared to amounts in soil. The uptake may occur through sorption of vapors from volatilization of PCBs or through root absorption and translocat-

tion. Greater uptake of the less chlorinated biphenyls than their more chlorinated counterparts can be due to their higher volatility in the former and to higher water solubility in the latter process.

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TABLE 2. PCBs (Aroclors) in soil and in vegetables grown in soil amended with contaminated sediment during 1981.

Aroclor	Soil	Beet roots			Beet leaves			Turnip roots			Turnip leaves		
		μg/kg	μg/kg	BF ^a	RBF ^a	μg/kg	BF	RBF	μg/kg	BF	RBF	μg/kg	BF
1248	80	15	.187	94	22	.275	69	30	.375	188	32	.400	200
1254	1,880	16	.008	4	94	.050	12	16	.008	4	40	.021	11
1260	14,440	35	.002	1	52	.004	1	20	.002	1	27	.002	1
Total	16,400	66			168			66			99		

^aBF is bioaccumulation factor calculated by dividing the amount (μg/kg) of an Aroclor in the plant part by its amount in the soil. RBF is the ratio of Aroclor 1248 or 1254 compared to 1260.

TABLE 3. PCBs (μg/kg) in vegetables grown in amended soil during 1982.

Aroclor	Turnips					Beans			
	Leaves			Roots		Leaves	Stems	Pods	Seeds
	Harvest date (1982)			Unpeeled	Peeled				
	7/31	8/4	8/12						
1248	27	31	20	32	3	44	37	44	13
1254	59	49	91	61	22	40	0	22	4
1260	52	79	156	204	10	107	36	98	0

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