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# BEHAVIOUR OF POLYCHLORINATED BIPHENYLS (PCBs) IN SOILS AND PLANTS

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## ABSTRACT

*A review of the literature indicates that polychlorinated biphenyls (PCBs) are not leachable in soils and that they are readily adsorbed by soil constituents. It appears that lower chlorinated PCBs are less adsorbed and thus slightly mobile in soils. Total organic matter content of soils seems to be more important than total clay content or total surface area in explaining adsorption of PCBs by soils. There have been few reports of the effect of PCBs on plants. These reports indicate that plants absorb PCBs, but in very low amounts. PCBs appear to have some effect on photosynthesis and respiration in plants.*

## INTRODUCTION

Numerous reviews on polychlorinated biphenyls (PCBs) have recently appeared and have been both general and intensive in their approach to the subject (Gustafson, 1970; Jensen, 1972; Peakall, 1972; Hutzinger *et al.*, 1974*b*; Klein & Weisgerber, 1976; WHO, 1976; Pomerantz *et al*, 1978; Pahren *et al.*, 1979; Pal *et al.*, 1980). The objective of this paper is to provide an intensive review of PCB behaviour in soils and in terrestrial higher plants. By necessity, due to gaps in information on certain aspects, we have included information from other areas of research, attempting to confirm its interpretation to the interest of this particular review.

The appearance of PCBs was first identified in Scandinavia in the aquatic environment and in eagles which feed on fish (Jensen, 1966; Jensen *et al.*, 1969). Since that time, alarm has been raised over the increasing appearance of PCBs in widespread areas of the aquatic environment. Findings of PCBs in terrestrial areas have also increased. Sewage sludges originating from several municipalities are

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reported to contain PCBs (Choi *et al.*, 1974; Furr *et al.*, 1976; Lawrence & Tosine, 1977; Bergh & Peoples, 1977; Pahren *et al.*, 1979) and industrial sources of PCB entry into the environment have been adequately documented (Nisbet & Sarofim, 1972). Although some recent surveys of urban and agricultural soils have reported little PCB contamination (Carey *et al.*, 1979*a*,*b*; Lang *et al.*, 1979), home gardens which had received dried sludge contained up to 50 ppm or greater (Bergh & Peoples, 1977) and certain Japanese agricultural soils contained up to 1200 ppm PCBs (Fujiwara, 1975). Plastic mulching is suspected as a source of PCB contamination in some Florida agricultural soils (Requejo *et al.*, 1979). Airborne transport has been suggested as a possible vector of movement to sites near a production facility (Stratton & Sosebee, 1976) and in one of the Great Lakes (Eisenreich *et al.*, 1979). This mode of transport as either fallout of particulate matter or in the vapour phase may explain the occurrence of PCBs in areas far from industrial sources of pollution (Keil *et al.*, 1972; Nisbet & Sarofim, 1972; WHO, 1976; Kalmaz & Kalmaz, 1979). The volatilisation of PCBs has been demonstrated to occur from a variety of surfaces and soils which lends credence to the possibility of airborne transport and fallout as a source of PCBs to the terrestrial and aquatic ecosystems (Oloffs *et al.*, 1973; Hutzinger *et al.*, 1974*b*; Mackay & Leinonen, 1975; Haque & Schmedding, 1976; Klein & Weisgerber, 1976; Kilzer *et al.*, 1979). One cannot entirely rule out completely natural sources of biphenyl formation by a variety of processes including fire, geological events and biosynthesis (Blumer & Youngblood, 1975). Plants themselves may form chemicals with a biphenyl structural bond (Borneff *et al.*, 1968; Cotterill *et al.*, 1974), although this natural source is likely to be insignificant when compared with other sources of biphenyl.

## POLYCHLORINATED BIPHENYLS IN THE SOIL

The mobility of chemicals in a particular soil depends in part on the soil properties and the chemical properties of the leaching substance. Soil properties which exert a considerable influence on the mobility of chemicals are: amount and types of colloids present, particle size distribution, pore volume and size distribution, water content, type of ground cover and slope (Weber & Weed, 1974). Chemical properties important to soil movement of chemicals are water solubility, ionisability, volatility and certain structural aspects or steric effects (Weber & Weed, 1974). It is important to remember that the soil system is a dynamic system involving a continuous interplay of combinations of these factors and is never at equilibrium.

PCBs are non-ionic compounds of extremely low water solubilities (Hutzinger *et al.*, 1974*b*). They are very lipophilic and are thus soluble in many organic solvents. Reported water solubilities range from 4130 to 0·016 pptm for single isomers (Weil *et al.*, 1974; Haque & Schmedding, 1976; Kilzer *et al.*, 1979) and from 15,000 to

2·7 pptm for Aroclors (Mackay & Leinonen, 1975; Pal *et al.*, 1980). Water solubility tends to decrease with increasing chlorination of the biphenyl ring (Fig. 1), but steric position of the chlorines also influences water solubility (Weil *et al.*, 1974; Wallnöfer *et al.*, 1975). Water solubility controls the amount of PCBs which dissolves in water; thus it is a very important property in assessing its movement in soils.

The vapour pressures of PCBs range from $7 \times 10^{-3}$ to $4 \times 10^{-5}$ mm Hg at 25 °C for Aroclors 1221–1260, which are in the low-to-moderate range, relative to other organic chemicals (Hutzinger *et al.*, 1974*b*). Increased chlorine content of the biphenyl generally results in decreased volatilisation. Loss of less chlorinated biphenyls from a sandy loam (0·6 % OM) occurred at a more rapid rate than for more highly chlorinated biphenyls when Aroclor 1254 was incorporated to a depth of 15 cm (Fig. 2). Chlorination was related to the relative peak position on a gas chromatograph, which follows a general trend of lower chlorinated isomers eluting earlier from the column (Lee *et al.*, 1979). Figure 2 shows that the lower chlorinated isomers exited more quickly from the soil. Peak 1 (lower chlorinated biphenyls) could not be recovered after 6 months, whereas 82 % of peak 7 (higher chlorinated biphenyls) was recovered after 8 months. Similarly, after 1 year of cropping a sandy loam, 79 % of chloroalkylene-9-$^{14}$C (isopropylated dichlorobiphenyls), 67 % of 2,4′,5-trichlorobiphenyl-$^{14}$C and 42 % of 2,2′,4,4′,6-pentachlorobiphenyl-$^{14}$C were vaporised from the top 1 cm of soil (Moza *et al.*, 1979*b*,*c*). Percentages are based on amount originally applied and illustrate greater loss of the lower chlorinated biphenyls under outdoor conditions. A criticism of these types of studies must include the fact that the amount of volatilisation was obtained by difference, not by analysis of PCB captured in the air (Pal *et al.*, 1980). However, since PCBs are slowly degraded and do not leach to an appreciable degree, the assumption that loss was due to volatilisation may be a safe one. Not only does chlorination influence volatility, but there also appears to be some evidence that steric factors may also prove important (Haque & Kohnert, 1976). The diffusion coefficient of 4,4′-dichlorobiphenyl is unusually low compared with that of 2,4′-dichlorobiphenyl. As may be expected, temperature also influenced the volatility of a PCB. A much

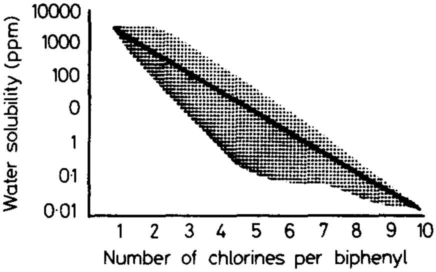

Fig. 1. Influence of chlorination on water solubility of biphenyl (redrawn from Weil *et al.* (1977)).

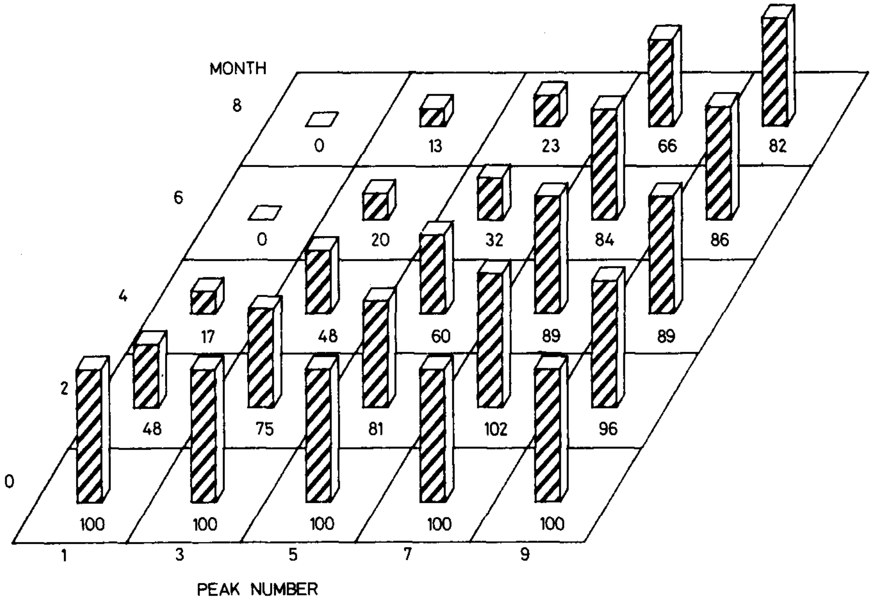

Fig. 2. Per cent recovery (in ppm equivalents) of gas chromatographic peaks of Aroclor 1254 from a Laveen loamy sand (drawn from data of Iwata *et al.* (1973)).

greater loss of Aroclor 1254 occurred at 60 °C than at 25 °C (Haque *et al.*, 1974). In these same series of experiments, Haque *et al.* (1974) found that greater loss of the PCB resulted from alternately wetting and drying a soil (cycled) than leaving it dry (Fig. 3), suggesting that water content may impart a considerable influence on mobility of PCBs in the soil. It could be expected that water could compete with PCBs for certain adsorption sites in the soil. The difference in loss due to the presence of water increased from almost no difference for tetrachlorobiphenyl to a twofold difference for heptachlorobiphenyl.

PCBs appear to volatilise much more readily from water than their vapour pressures would indicate (Oloffs *et al.*, 1972, 1973; Mackay & Wolkoff, 1973), prompting the suggestion that hydrophobic organics accumulate at the water–air interface, which may result in an even more volatile behaviour of the chemicals (Oloffs *et al.*, 1972; Spencer *et al.*, 1973). Volatilisation from water of 2,4′,5-trichlorobiphenyl reached 43 % and 2,2′,4,4′,6-pentachlorobiphenyl reached 36 % in 2 h, yet only 3·5 % of biphenyl (no chlorination) evaporated within 2 h under the same conditions (Kilzer *et al.*, 1979), despite the high vapour pressure reported for biphenyl (Davis & Munroe, 1977). The water solubilities of the biphenyl, tri- and pentachlorobiphenyl were given as 6000–7500, 110 and 31 pptm, respectively. The solubility limit of the pentachlorobiphenyl was exceeded. One would expect that additional chlorines would add weight to the molecule and lower the capacity to

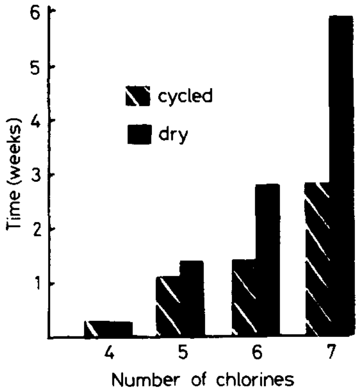

Fig. 3. Time required for the disappearance of 20% of applied Aroclor 1254 from cycled (watered daily) and dry sand (drawn from data of Haque *et al.* (1974)).

vaporise from water as it does in soils, but the behaviour appears to be quite opposite. The answer may lie in interpretation of the water solubilities. Since biphenyl was the most water-soluble compound, it had a greater affinity for the water, and this may decrease its tendency to escape the water, whereas the highly hydrophobic PCBs possess such low affinity for water that they readily accumulate at interfaces. Mackay & Leinonen (1975) reported that Aroclors 1242–1260 possessed higher partial pressures in water. Notwithstanding the previous criticism of volatilisation studies, these data are likely still to be useful in predicting trends relevant to PCB behaviour in the environment.

Despite the apparent increase in mobility of PCBs in wet soils, the amounts of PCBs leached through soils have been reported to be extremely low. No detectable chloralkylene-9 (a mixture of propylated dichlorobiphenyls), trichlorobiphenyl or pentachlorobiphenyl leached from 60 cm deep boxes of a sandy loam soil after 1 year of outdoor cropping (Moza *et al.*, 1979*b*). During the second year, 0·1, 0·2 and 0·1% of the originally applied activities of the di-, tri- and pentachlorobiphenyl, respectively, were collected as leachate. Rainfall over the 2-year period totalled 406 mm. This low amount of PCB may have escaped the boxes by being attached to soil colloids washing through drain holes in the box bottoms. Barely detectable (less than 0·01%) radioactivity was analysed leaching from 7·6 cm of a Lakeland sand treated with 0·5 μCi (20 ppm) total of a PCB mixture of Aroclor 1254 and labelled material resembling Aroclor 1254 (Strek *et al.*, 1981). A mixture of PCBs (containing an average of 2·7% chlorines per molecule) was surface-applied to an acidic, sandy forest soil, and soil probes were taken to 85 cm in depth with the aid of a mechanical borer (Scharpenseel *et al.*, 1977*b*). Unfortunately, because the data were not reported in disintegrations per minute or per cent of applied material, no significance can be attached to the proposed PCB levels in the lower profiles.

Leaching studies using soil columns provide a more controlled environment than measuring leaching from containers. Data collected in this manner can also provide useful information for predicting trends in the field situation. Tucker *et al.* (1975) used three soil types and applied 25,000 ppm Aroclor 1016 to 7·6 cm of soil in a column, placing it above 7·6 cm of untreated soil. Soil types included Norfolk sandy loam (5·5 % clay, 1·0 % OM), Ray silty loam (9·6 % clay, 1·0 % OM) and Drummer clay loam (35·8 % clay, 6·0 % OM). Flow rates, in respective order for the soils, were 0·26, 0·53 and 0·32 litres day$^{-1}$ until totals of 48·1, 98·1 and 59·2 litres, respectively, were collected. Despite the high leaching rates and the unusually high application rate of 25,000 ppm, only a maximum of 0·05 % of that applied was leached through 7·6 cm of soil. The highest rate of loss, from the Ray silty loam, can be explained as a result of the relatively higher flow rate. Tucker *et al.* (1975) attributed the apparent high affinity of Aroclor 1016 for the clay loam soil to the clay content. The organic matter content also happened to be the highest of the three soils, which might provide a better explanation. In another leaching study, 50 μCi of a mixture of chlorobiphenyls (containing an average of 2·7 % chlorines per molecule) were applied to *in situ* cores of an Altvega loessal soil; only 0·442 and 0·725 % of the applied radioactivity was leached through the 50 cm column of soil following application of 900 and 1700 ml of water (Scharpenseel *et al.*, 1977*a*). It appeared that the vertical movement was fairly high in this experiment. The use of undisturbed soil cores, which may have contained natural channels, and the possibility of boundary flow (channelling down the side) may, in part, account for the PCB movement which did occur. These considerations have been deemed important in the study of herbicide mobility in soils (Weber & Peeper, 1977). In both the previous experiments, chlorination of the biphenyl was medium to low (averaging 42 and 27 %, respectively) and leaching of the PCBs was extremely low. It could be expected that, due to their lower solubilities, higher chlorinated biphenyls may not be as mobile in soils. Hexa- and tetrachlorobiphenyl adsorbed onto a Woodburn soil to a greater degree than did dichlorobiphenyl (Fig. 4), suggesting a higher affinity of higher chlorinated biphenyls to soils. Suzuki *et al.* (1977) and Tucker *et al.* (1975) found that lesser chlorinated biphenyls leached more than higher chlorinated biphenyls under similar conditions, possibly due to their greater water solubilities. PCBs would not appear to move readily through the soil profile. Consequently, there have been few reports of PCBs in groundwater (Lee *et al.*, 1979) and, even then, contamination cannot be directly attributed to leaching. The overwhelming evidence shows that PCBs do not leach in soils to any significant degree under normal conditions and that they possess a strong affinity for the soil. Little is known about the soil behaviour of the minute amounts of PCB hydroxy and methoxy metabolites which may result from impurities in the commercial mixtures, or from PCB degradation. These compounds, because of their higher water solubilities, may be more mobile than the original parent PCB compound.

Because of their strong affinity to soil particles, chlorinated hydrocarbons of low

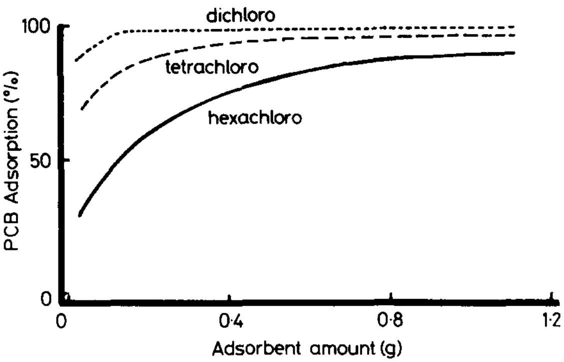

Fig. 4. Relative adsorption of PCB isomers onto Woodburn soil as a function of chlorine content (redrawn from Haque & Schmedding (1976)).

water solubility can sometimes move in soil profiles under certain conditions. Ballard (1971), in a discussion of the movement of DDT—another chlorinated hydrocarbon of low water solubility (0·001 ppm)—through a forest soil, discovered that 91 % of the applied *o,p'*-DDT was recovered in the humic acid fraction and only 9 % in the fulvic acid fraction of the soil organic matter. Fulvic acids are generally more oxidised and acidic than humic acids and thus more water soluble (Weed & Weber, 1974) and would not offer a compatible surface for the adsorption of PCBs and other hydrophobic organics. Alternately, humic acids are more lipophilic in nature, insoluble in water (except at extremely high pH) and could be expected to adsorb a higher proportion of PCBs. Ballard suggested that for DDT, its mobility in soils is controlled by the solubility of the carrier material (humic acid), an idea that may be extrapolated to PCBs. Scharpenseel *et al.* (1977*a*) found a close association of radiolabelled PCBs with the organic component of a soil. Separation of the organic fraction by the classic method of alkali extraction demonstrated a more favourable association of the PCBs with humic acid. A similar relationship was described by Moza *et al.* (1979*b*) who separated the organic fractions of a soil which had been previously subjected to a hot methanol extraction into humin, humic acid and fulvic acid. More of the PCB was associated with the humin (extremely insoluble in water) than with any other fraction, but adsorption was dependent on the specific isotope. It may have been more revealing to separate and analyse the fractions prior to methanol Soxhlet extraction. Ballard (1971) warned that a great increase in pH in an organic soil may cause the solubilisation of the humic substances and may result in increased leaching of the carrier material and, with it, the attached chemical. PCBs may behave in a similar manner due to the similarity of their chemical properties to DDT. Erosion or other processes may cause movement of the substrate to which PCBs are attached. This may represent the major vector of movement of PCBs from land into open water. There is no evidence to indicate that PCBs have leached in appreciable amounts and the majority probably remain relatively near to their sites of application, slowly degrading to form intermediary metabolites.

Adsorption studies using organic chemicals in aqueous solution have explored the relationships between the chemical and soil colloids. The greatest limitation in using PCBs in aqueous adsorption studies is the extremely low water solubilities of the chemicals. The solubility limit must not be exceeded for the correct interpretation of adsorption isotherm studies. The moderate volatility of PCBs in water must also be taken into consideration. The importance of organic matter in PCB adsorption in soils has already been pointed out. Studies using aqueous Aroclor 1254 and sand, silica gel, various clays and a silty loam soil (3·1 % OM, 16·6 % clay) as adsorbents showed a greater affinity of the PCB for the soil, explained by the authors as directly resulting from the organic matter content of the soil (Haque *et al.*, 1974). Haque & Schmedding (1976) demonstrated that humic acid was an extremely effective adsorbent of di-, tetra- and hexachlorobiphenyls, more so than a silty loam soil, illite clay or a quartz sand. When aqueous Aroclor 1254 was adsorbed to activated carbon (100 % OM), a peaty muck (92·2 % OM), montmorillonite clay (1·6 % OM), Lakeland sand (1·4 % OM) and peroxide-treated Lakeland sand (0·2 % OM), adsorption was highest in the activated carbon and decreased with decreasing organic matter content (Fig. 5). Lee *et al.* (1979) found that Freundlich adsorption coefficients (*K*'s) of Aroclor 1242 on several adsorbents were highly linearly correlated ($r^2 = 0{\cdot}87$) with total organic content (Fig. 6). Adsorbents used in this study included medium temperature coal char

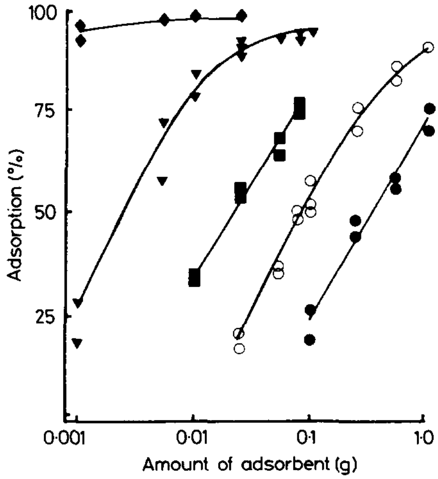

Fig. 5. Adsorption of a $^{14}$C-PCB from aqueous solution by activated carbon (◆), organic matter (▼), montmorillonite clay (■), Lakeland sand (○), $H_2O_2$ treated Lakeland sand (●). (From Strek & Weber (1982)).

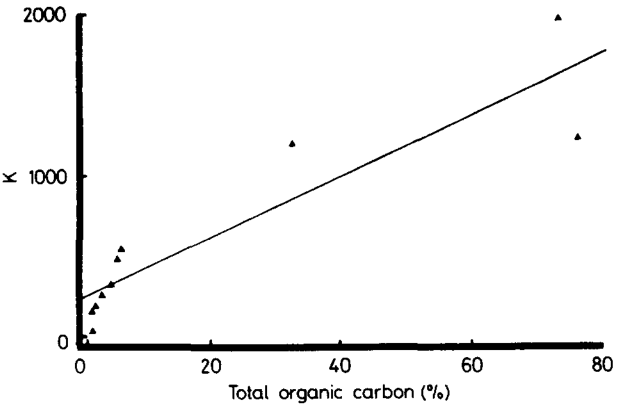

Fig. 6. PCB (Aroclor 1242) adsorption constant ($K$) as a function of total organic carbon content (%) of soil and soil-like materials (redrawn from Lee *et al.* (1979)).

(74·04 % OM) and high temperature coal char (76·62 % OM)—both low grades of activated carbon—Catlin silty loam (4·73 % OM), montmorillonite clay (0·93 % OM) and Ottawa sand (<0·01 % OM), listed in order of greater to lower relative adsorption of the PCB. They also observed a trend of greater adsorption of the more highly chlorinated isomers, noted previously by Haque & Schmedding (1976) and, in addition, they pointed out that position of the Cl on the biphenyl ring influenced relative adsorption. The desorption of PCBs has been reported to be both rapid (Steen *et al.*, 1978) and negligible (Haque *et al.*, 1974; Hiraizumi *et al.*, 1979); however, the evidence favours the idea that PCBs are not readily desorbed from surfaces by water, again most probably due to their highly hydrophobic properties.

The importance of surface area to PCB adsorption is apparent when attempting to work with the chemical in aqueous solutions. Numerous workers have reported that PCBs readily cling to surfaces (Zitko, 1971; Gresshoff *et al.*, 1977; Lee *et al.*, 1979). Using Kaneclor 500 and aged activated carbon, activated sludge, seabed sediment, sand and zooplankton, Hiraizumi *et al.* (1979) described an increasing relationship between PCB concentration factor and specific surface area ($N_2$ method) of the adsorbents (Fig. 7). The authors delegated only secondary importance to organic matter content. This interpretation conflicts with that of Lee *et al.* (1979), who also measured surface area with $N_2$, but found no correlation to the Freundlich adsorption coefficient $K$. These two lines of data cannot be compared directly, for Hiraizumi *et al.* (1979) used concentration factor and Lee *et al.* (1979) used $K$ to plot against specific surface area. This leaves the divergence of interpretation partly open to debate. Table 1 shows the relative adsorption of aqueous Aroclor 1254 by various adsorbents using the amount of adsorbent needed to adsorb 50 % of the available PCB from solution ($A_{50}$) as an indicator for

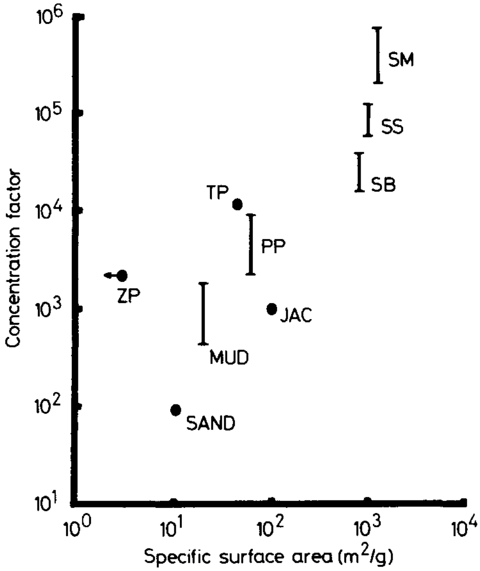

Fig. 7. Relationship between PCB concentration factors and specific surface area ($m^2 g^{-1}$) of adsorbing agents. ZP, zooplankton; SAND, Iwafume coarse sand; MUD, seabed mud; TP, total plankton; PP, phytoplankton; JAC, Japanese acid clay; SB, SM, SS, suspended matter in bottom, middle, and surface layer, respectively (redrawn from Hiraizumi *et al.* (1979)).

adsorption. Had the $N_2$ surface area been most important to adsorption, the montmorillonite would have been the most effective adsorbent since its surface area was the largest. Yet, the peaty muck was sixteen times more effective. The increase in PCB adsorption between the peaty muck and montmorillonite may express a qualitative difference in adsorption mechanism, perhaps a chemical complexing by the organic matter (Strek & Weber, 1982). Although its surface area was low ($1{\cdot}23\,m^2 g^{-1}$), the $N_2$ method cannot measure the interstitial surfaces of organic matter expected to be available for adsorption, which should largely increase the effective surface area. Lee *et al.* (1979) surmised that PCBs did not adsorb into the interlayer of montmorillonite, a reasonable assumption due to the extremely hydrophobic nature of PCBs. However, the increase in outer surface area may be responsible for the fourfold increase in adsorption by the montmorillonite over the Lakeland sand, despite their similar organic matter contents (Table 1, Fig. 5). Surface area obtained by $CO_2$ adsorption correlated linearly with organic content (Lee *et al.*, 1979); however, total organic carbon content was more important by a factor of three. The use of $CO_2$ surface area measurements is perhaps an indication of the amount of carbon surface because of the availability of aldehydic, carboxylic and carbonylic groups to which it can physically bond. On a practical level, total

TABLE 1
PROPERTIES OF SOILS AND ADSORBENTS

| *Adsorbent* | *Organic matter*[a] (%) | *Specific surface area*[b] ($m^2 g^{-1}$) | $A_{50}$[c] (*g*) | *Relative ranking of adsorptivity* |
|---|---|---|---|---|
| Activated carbon | — | 452·18 | — | — |
| Peaty muck organic matter | 92·2 | 1·23 | 0·0025 | 132·0 |
| Montmorillonite | 1·6 | 62·97 | 0·0210 | 16·5 |
| Lakeland sand | 1·4 | 0·41 | 0·0800 | 4·1 |
| $H_2O_2$ treated Lakeland sand | 0·2 | 0·62 | 0·3300 | 1·0 |

[a] Organic matter determined by ashing at 400 °C.
[b] Specific surface area determined by $N_2$ adsorption (Quantachrome Quantasorb Surface Area Analyser).
[c] Amount of adsorbent necessary for 50 % adsorption of PCB using equilibrium concentrations of 0·000068 to 0·000063 mg $g^{-1}$.

organic carbon level is more easily obtained and seems to be more important to PCB adsorption.

The analysis of concentrations of PCB found in sediments can also prove revealing. The chlorinated hydrocarbon contents of harbour sediment samples were found to be linearly correlated to total organic carbon content (Choi & Chan, 1976). The partition coefficients of two tetrachlorobiphenyls correlated well with the organic matter content of a series of sediments; whereas, for Aroclor 1254, only a general increase in partition coefficient with organic matter content was noted (Steen *et al.*, 1978). Pavlou & Dexter (1979) also related PCB levels in offshore sediments to natural organic matter content.

Plant studies also indicate that organic matter content of soil influences the bioavailability of PCBs. Strek & Weber (1982) showed that additions of montmorillonite at 2·5 and 5·0 % were not as effective as peaty muck at the same levels in reducing the toxicity of 200 ppm Aroclor 1254 to pigweed (Fig. 8). At the 10 % level of addition, both adsorbents were equally effective in offering protection. Because activated carbon is such a good adsorbent of PCBs (Lee *et al.*, 1979; Weber & Mrozek, 1979; Shea *et al.*, 1980; Strek *et al.*, 1981; Strek & Weber, 1982) and other non-ionic organic chemicals (Lichenstein *et al.*, 1968; Mattson & Mark, 1971) and because it normally has an available surface area greater than soil organic matter, it would be expected greatly to limit the mobility of PCBs in soil. Activated carbon applied to soil eliminated the toxicity of soil-applied Aroclor 1254 to soybeans (Weber & Mrozek, 1979) and sugar beets (Strek *et al.*, 1981). Sugar beets that would not grow at 1000 ppm did so upon addition of 3333 ppm activated carbon to the soil. No differences in height, top fresh weight, and water use from control plants were noted in the carbon treatment. Protection by carbon from reduction of height, top fresh weight and water use in soybeans caused by PCB were also reported. The rate of activated carbon addition was only 3·3 times greater than the PCB, much less than the 100 to 500 safety factor recommended for activated carbon in treating

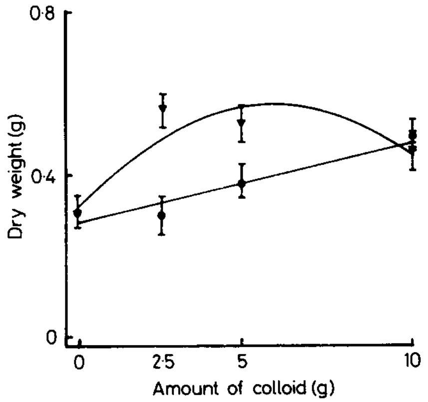

Fig. 8. Influence of montmorillonite clay (●) and organic matter (▼) amendments on toxicity of 200 ppm Aroclor 1254 to pigweed (from Strek & Weber (1982)).

problems with pesticides. The activated carbon treatment also reduced the translocation of $^{14}$C-PCB activity to the tops of beet, sorghum, peanut and corn by 81 to 100 %, illustrating the tenacity of adsorption and the reduction in bioavailability of Aroclor 1254 by activated carbon.

Most investigators propose a physical mechanism of adsorption for non-ionic chemicals onto organic matter because adsorption can be described by a Freundlich isotherm. Lee *et al.* (1979) showed that heats of adsorption, ranging from 5 to 18 kcal mole$^{-1}$, were well within the values ascribed to physical adsorption. Possible adsorption mechanisms include electrostatic attraction of the chlorines to surfaces, hydrophobic bonding, or perhaps Pi complexing of the biphenyl ring to other Pi orbitals in ring structures such as those found in organic matter.

## EFFECTS OF POLYCHLORINATED BIPHENYLS ON PLANTS

Inhibition of plant growth due to effects by PCBs has been documented mainly for algae. Substantial reductions in algal cell number have been noted at generally low levels (0·3 to 10 ppm) of PCBs in aqueous solution (Keil *et al.*, 1972; Cole & Plapp, 1974; Glooschenko & Glooschenko, 1975; Larsson & Tillberg, 1975; Ewald *et al.*, 1976; Gresshoff *et al.*, 1977; Bryan & Olofsson, 1978). Reports of growth inhibition to higher plants have been scarce. Mahanty & Fineran (1976) reported the complete internal disorganisation of chloroplasts in the frond cells of an aquatic plant *Spirodela oligorrhiza* (Kurtz) Hegelm. exposed to 5 ppm Aroclor 1242 Weber & Mrozek (1979) reported malformations on newly developed soybean leaves on plants growing in 1000 ppm Aroclor 1254 applied to soil. Reduction in plant heights and fresh weights were noted for soybean, beets and pigweed *Amaranthus* *retroflexus* L. (Fig. 9), but only fresh weight reductions were reported for fescue (Weber & Mrozek, 1979; Strek & Weber, 1980, 1982; Strek *et al.*, 1981). At the 1000 ppm rate of Aroclor 1254, soybean growth (in the second crop) was inhibited by 47 % (Fig. 9). Cumulative water use seems to be more sensitive than plant growth to PCB (Strek *et al.*, 1981), which may also mean that effects on plant growth may be indirect, following effects which reflect on transpiration.

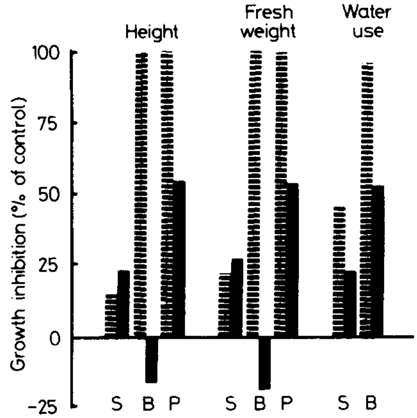

Fig. 9. Effect of 1000 ppm (striped bars) and 100 ppm (black bars) of soil-applied Aroclor 1254 on soybean (S), beet (B) and pigweed (P) growth (drawn from data of Strek & Weber (1980) and Strek *et al.* (1981)).

Uptake into plants is possible through two general routes. One route is through the root system and the other is through prior adsorption to the foliage and stems and subsequent movement through the epidermal layers into the apoplast or symplast. The former route is probably the most important way of uptake for soil-applied PCBs, while the latter route probably predominates in the uptake of airborne PCBs by terrestrial plants and dissolved PCBs by aquatic plants and microorganisms. However, uptake of PCBs from fallout is unlikely to occur to any great degree because the chemical may merely adsorb to the outer surfaces of the plant and may not be truly present inside the plant. The cuticle contains many lipophilic compounds in which the PCBs could effectively 'dissolve', limiting further internal migration. In addition, unless PCB uptake by microbes can be differentiated into that which has adsorbed to the surface and that which has entered the protoplasm proper, uptake studies of this nature (using algae or bacteria) will become misinterpreted. Uptake of $^{14}$C-labelled PCBs following application to leaves has been demonstrated, although in low amounts (3·2 to 15·5 %) of that applied; the greatest loss probably occurred through volatilisation (Moza *et al.*, 1976*b*; Weber & Mrozek, 1979). Plant uptake from the soil has also been demonstrated and will be discussed below.

Statistical analyses were performed on plant PCB uptake data taken from several sources (Iwata *et al.*, 1974; Wallnöfer & Königer, 1974; Wallnöfer *et al.*, 1975; Iwata & Gunther, 1976; Moza *et al.*, 1976*b*, 1979*a*,*b*,*c*; Weber & Mrozek, 1979). The data of Lawrence & Tosine (1977) were not included because they were reported in units of $\mu$g litre$^{-1}$. The data of Iwata *et al.* (1974) and Iwata & Gunter (1976) were reported in ppm equivalents of Aroclor 1254 for several chromatographic peaks, which were averaged for the five peaks reported. Only those data for which the plants were grown on soils treated with PCBs were included. Input data included isotope, number of chlorines per biphenyl, plant part analysed, PCB content in ppm, PCB content of the soil at planting and at harvest and growth period. Species included carrot, fescue, radish, soybean, spruce, sugarbeet, tomato and unidentified weeds. The species were planted in soils fortified with 0·046 to 100 ppm and at harvest soil levels ranged from 0·040 to 76 ppm. Surprisingly, several factors proved to be highly significant and certain trends could be distinguished despite the wide range of experimental conditions. The factors—species, degree of chlorination of biphenyl and growth period gave the most significant explanation of plant uptake. Amounts of uptake were generally low, ranging from 0·0016 to 13·9 ppm, and averaged 1·241 ppm over all data.

The data suggested that PCB content of the plant was dependent on the PCB concentration in the soil. The PCB concentration in the plant was analysed versus soil concentrations at both planting and at harvest and was found to be significant ($PR > F \leq 0{\cdot}0001$) for both soil sampling times. Since PCB availability and mobility in soils are greatly influenced by clay and organic contents, as discussed previously, as well as temperature (Haque *et al.*, 1974), one must consider these factors as well when determining whether levels in a soil are high enough for potential uptake.

Differences between plant species were also significant, with carrots containing an average amount of 2·52 ppm, fescue averaging 1·67 ppm and unidentified weeds averaging 1·52 ppm. No common basis for comparison existed; thus no sound interpretations of these data are possible. Tomatoes contained the least amount, averaging 0·0023 ppm, the low content probably a result of the application of extremely low rates of PCB of between 0·046 to 0·062 ppm (Wallnöfer & Königer, 1974). Carrots are described as accumulators of lipophilic chemicals and would be expected to take up relatively more PCBs than other crops (Lichtenstein *et al.*, 1968; Iwata *et al.*, 1974; Moza *et al.*, 1976*b*). Carrots contained the highest average PCB content of all species (2·52 ppm). There appears to be a limit of PCB concentration in the soil at which no detectable PCB can be taken up by plants. Wallnöfer *et al.* (1975) could not detect any 4,4′-di-, 2,2′,5,5′-tetra-, or 2,2′,4,4′,5,5′-hexachlorobiphenyl applied to soil at 0·05 ppm in carrots grown for 120 days. This could have been due to the physical bonding of PCBs to the soil colloids and consequent lesser mobility and availability. Soil colloids can substantially reduce the toxicity of PCBs to plants. The addition of 5 % organic matter to Lakeland soil reduced the fresh weight inhibition of 200 ppm soil-applied Aroclor 1254 to pigweed from 49 % to

8 %; addition of 5 % montmorillonite clay only reduced fresh weight inhibition from 49 % to 31 % (Fig. 8).

Plant roots (carrot and sugarbeets) averaged 2·02 ppm PCBs in the statistical survey, whereas plant tops averaged only 0·52 ppm PCBs, but the difference between the parts did not prove to be statistically significant. Perhaps this was due to the large range in PCB content from treatments in which very low amounts of PCBs were applied, and because only the PCB content of plant tops was reported for these studies. Iwata *et al.* (1974), Iwata & Gunther (1976) and Wallnöfer *et al.* (1975) analysed carrot root peelings and reported that 97 to 100 % of the detectable PCBs were contained in them. This finding suggests that the roots of many plants could have what is considered to be 'absorbed' PCB simply adsorbed to the exterior surface. PCBs found 'in' carrot roots and possibly other plant roots may not have penetrated the root at all, but merely adsorbed to the outer surface, and analysis of the entire root reflects PCB contained both internally and on the exterior surfaces of the root. This possibility of adsorption to the outer root surface was also suggested by Pahren *et al.* (1979). Because of their low water solubility and high lipophilicity, PCBs are expected to move into plants via the symplastic route. More research is needed to determine the exact route of movement.

Amount of chlorination of the biphenyl has been suggested as being an important factor in PCB uptake by plants and it appears that PCBs of lower chlorination are taken up much more readily that those of higher chlorination (Iwata *et al.*, 1974; Wallnöfer *et al.*, 1975; Moza *et al.*, 1976*b*; Suzuki *et al.*, 1977). However, per cent chlorination of biphenyl over all data entries as a factor in PCB uptake showed no significant differences and no distinguishable trend in the statistical survey. This may have been caused by the inclusion of data in which commercial PCB mixtures were applied. Chlorination of Aroclor 1254 ranges from 0 to 7 per molecule, with a greater majority containing four to six chlorines (Pal *et al.*, 1980). When the Aroclor 1254 (a commercial mixture) data are ignored in the statistical survey, there follows a general trend of increasing PCB content with decreasing chlorination. When plant PCB content versus per cent chlorination and plant part were analysed, both main effects as well as the interaction effect, were highly significant, suggesting statistically that the amount of chlorination affected the mobility of PCBs within a particular plant part. Since lower chlorinated PCBs have been reported to be more mobile in soil than highly chlorinated PCBs, they may be more readily transported and available for plant uptake. Strek *et al.* (1981) reported decreases in toxicity of Aroclor 1254 to soybean and translocation of $^{14}C$ activity, applied as a PCB similar to Aroclor 1254 in composition, into fescue with continued cropping and harvest, perhaps an indication of the depletion of the lower chlorinated PCBs from the soil or fixation of PCBs by soil in non-biologically available form.

Early work on PCB inhibition of photosynthesis was performed using algae. Inhibition of batch photosynthesis in algae has been explored by many researchers (Cole & Plap, 1974; Glooschenko & Glooschenko, 1975; Ewald *et al.*, 1976;

Gresshoff *et al.*, 1977; Bryan & Olofsson, 1978). Fisher (1975) found a reduction in batch photosynthesis, but not on a per cell basis, relating the apparent reduction in photosynthetic rate to the reduction in cell quantity. Sinclair *et al.* (1977) cited difficulties with interpretation of photosynthesis inhibition data using algae as experimental species and the measurement of oxygen evolution by the cells. They declared that a reduction in oxygen evolution may be the result of an effect on respiration as well as photosynthesis. Another problem may be that effects on permeability may alter results based on movement of products across membranes (Sinclair *et al.*, 1977; Bryan & Olofsson, 1978). Ewald *et al.* (1976) noted reduced chlorophyll levels in *Euglena gracilis* following exposure to 4·4 ppm Aroclor 1221 for 48 h. Little research has been done to confirm the presumed effect of PCBs on photosynthesis in higher plants. Mahanty & McWha (1976) observed a characteristic 'striping' of alternate green and etiolated bands on fronds of an aquatic plant *Spirodela oligorrhiza*. They also reported reductions in chlorophyll and RNA contents of 56 and 27 %, respectively, in plants grown in 5 ppm Aroclor 1242, but no reduction in DNA levels could be detected. Using transmission electron microscopy, Mahanty & Fineran (1976) concluded that PCB apparently stimulated the conversion of chloroplasts to chromoplasts, resulting ultimately in their complete disorganisation. They found no detectable damage to the double membrane, in contrast to the observed distortion of the chloroplast lamellae in algal cultures treated with various Aroclors (Glooschenko & Glooschenko, 1975). Veinal chlorosis of terminal leaves of soybean on plants grown in 1000 ppm Aroclor 1254 was observed by Weber & Mrozek (1979) and Strek *et al.* (1981). Sinclair *et al.* (1977) reported a 60 % reduction in oxygen evolution from isolated spinach chloroplasts at both 10 and 20 °C using $10^{-4}$ M Aroclor 1221. The inhibition occurred even in the presence of an uncoupler, and they found no inhibition of PS I when PS II was blocked, leading to the conclusion that Aroclor 1221 inhibits PS II.

Most of the work done on the effect of PCBs on respiration has also been performed using algae (Larsson & Tillberg, 1975; Ewald *et al.*, 1976; Sinclair *et al.*, 1977), as well as rat liver mitochondria (Sivalingan *et al.*, 1973) and bacteria (Blakemore & Carey, 1978). The effect of PCBs on respiration appears to be a matter of debate, with reports showing both inhibition (Sivalingan *et al.*, 1973; Larsson & Tillberg, 1975; Sinclair *et al.*, 1977) and lack of inhibition (Blakemore & Carey, 1978; Ewald *et al.*, 1976) occurring for various PCBs at differing concentrations. Where inhibition of respiration has been reported, the behaviour of PCB as an uncoupler is mentioned. Sivalingan *et al.* (1973) related lower toxicity to rat liver mitochondria with lower chlorination of the biphenyl, in contrast to the generally higher toxicity of lower chlorinated biphenyls to *Aspergillus flavus* cultures found by Murado *et al.* (1976). Larsson & Tillberg (1975) refer to the possibility that inhibition of respiration may be due to structural changes in the mitochondrial membrane, a criticism that is similar to those of photosynthesis studies using algae.

The importance of biphenyl metabolites in plants has often been overlooked. In a variety of plants, the main metabolites which have been isolated appear to be mono- and dihydroxylated biphenyls (Moza *et al.*, 1973, 1976*a*,*b*, 1979*a*,*b*,*c*). Additionally, methoxy derivatives have been isolated (Moza *et al.*, 1976*b*, 1979*b*). Unidentified conjugates also seem to be important metabolites (Moza *et al.*, 1973, 1976*a*,*b*, 1979*a*,*b*,*c*). Microbial breakdown yields many products, including phenols and benzoic acids (Pal *et al.*, 1980). There has even been one report of the dechlorination of a hexachlorinated biphenyl in rabbit (Hutzinger *et al.*, 1974*a*), although soil microorganisms appear to lack such ability (Pal *et al.*, 1980). In their review, Klein & Weisgerber (1976) discuss metabolites and mechanisms of hydroxylation for plants and animals. The phenolic and oxide metabolites of tetrachlorobiphenyls have been shown to possess higher toxicity to animal cells (Yamamoto & Yoshimura, 1973; Stadnicki & Allen, 1979). Chlorinated dibenzofurans and dioxins, of much higher toxicity than chlorinated biphenyls (Pomerantz *et al.*, 1978), have been isolated from a variety of commercial PCB mixtures (Bowes *et al.*, 1975; Pomerantz *et al.*, 1978). Although this presumption has not been tested on plants, impurities in PCBs may be more toxic than the parent compound itself (Hammond *et al.*, 1972). This idea may also be applicable to the plant-manufactured hydroxylated metabolites themselves which may be causing the various detrimental effects attributed to the PCBs. More studies are required before valid trends can be discovered.

## SUMMARY

The mobility of polychlorinated biphenyls (PCBs) in the terrestrial ecosystem depends on their chemical properties, the properties of the soil in which they are present and the characteristics of the plants exposed to them. Due to their non-ionic nature, high lipophilicity, generally low volatility, low water solubilities and ability to resist degradation, PCBs are expected to persist in the environment.

Although vapour pressures of PCBs are moderate to low compared with other pesticides, they can escape from soils. Lower chlorinated biphenyls appear to volatilise more rapidly, although the position of the chlorines exerts some influence. Higher temperature induces a higher loss of PCBs from the soil. PCBs seem to volatilise from water and from soil at a much higher rate than their vapour pressures would suggest. The hydrophobic properties of PCBs may contribute to this phenomenon by causing accumulation of the PCBs at the water–air interface, which may result in quicker loss. PCBs possess a high affinity for the soil and do not leach significantly. Higher chlorinated isomers are generally less mobile and more readily adsorbed, but this also depends upon stereochemistry. Organic matter seems to be the most important soil constituent to PCB adsorption, but specific surface area is also related, and this suggests that other soil components also bind PCBs.

PCBs appear to adsorb more to the humin fraction than to the humic and fulvic acid fractions of the soil organic matter. Relationships between PCB adsorption parameters and per cent organic carbon and surface area, as measured by $N_2$ and $CO_2$, have been described. Investigators support a physical mechanism for adsorption of PCBs to soil constituents, but chemical complexing may prove a likely mechanism.

Effects of PCBs on plant growth can appear as reductions in height, weight and water use. Malformations of growth and chlorosis occur in soybeans only at high rates of application. A characteristic striping pattern of *Spirodela oligorrhiza* has been observed, as well as internal disorganisation of chloroplasts. Uptake into plants has generally been low, reports ranging from 0·00016 to 13·9 ppm and averaging 1·241 ppm. Carrots seem to have the ability to accumulate PCBs in the highest amounts, although high PCB content of the roots may be attributed primarily to surface adsorption. Lower chlorinated biphenyls appear to be translocated by plants in higher quantities than highly chlorinated biphenyls. Length of growth period can affect plant PCB content, but may not be as important as PCB content in the soil. PCBs can reduce chlorophyll content and may affect photosynthesis, possibly at PS II, and may also uncouple respiration; however, effects on membrane permeability may be confounding these results. Not much is known about the movement of trace amounts of hydroxy metabolites in soils, nor the effects of these metabolites on plants. The hydroxy metabolites of PCBs in plants and soils and impurities present in commercial PCBs may ultimately be responsible for the toxic effects ascribed to the parent compound.

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