Bull. Environm. Contam. Toxicol. 27, 689–694 (1981)

Uptake of Polychlorobiphenyls Present in Trace Amounts from Dried Municipal Sewage Sludge Through an Old Field Ecosystem

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Municipal sewage wastes are increasingly being used as fertilizers and soil conditioners. FURR et al. (1976) report that several major municipalities are already using sludge in this capacity. The cities of Chicago, Milwaukee, and Schenectady commercially market sludge under the tradenames Vertigreen, Milorganite, and Orgro, respectively.

Although the use of sludge as a fertilizer or conditioner is attractive in terms of cost, nutrient recycling, and waste disposal, a question has arisen regarding pathogens and trace levels of organic and inorganic toxic materials contained in the sludge, that may affect human health or the ecosystem where the sludge is eventually disposed (FURR et al. 1976; TORREY 1979). In particular, dried sewage sludge has been found to contain polychlorinated biphenyls (PCBs) at the ppm level.

This study examined PCB concentrations in a sewage sludge used as a fertilizer, in the treated soil, in the plants growing in that soil, and in arthropods in contact with the soil and feeding upon the plants. By repeated sampling of each level, the uptake of PCBs through a terrestrial food chain could be proposed.

MATERIALS AND METHODS

The research was performed at the Miami University Ecology Research Center, two miles north of Oxford, OH. Four acres (1.6 ha) of grassland, previously enclosed by 20 gauge galvanized steel walls extending 61 cm above the ground and 46 cm below the ground, were subdivided into 16 0.1 ha enclosures. In previous experiments at this site, cross contamination of pesticides and fertilizers had been negligible.

Five replicate plots were randomly selected for treatment, of which three were sludge-treated and two, serving as controls, received no treatment. Soil particle size analysis by the method of FOTH & TURK (1972) averaged 30.0% sand, 48.2% clay, and 21.8% silt. The soil samples averaged 5.9% organic matter. The plots were last planted in 1974 with a mixture of Kentucky Bluegrass (Poa pratensis), Rye grass (Lolium perenne), and Fescue (Festuca elatior). In 1979, Goldenrod (Solidago rigida) and the fescue comprised over 95% of the plant biomass present.

0007-4861/81/0027-0689 $01.20

The sludge used was "Milorganite," a dried, granular, processed sludge marketed by the Sewerage Commission of Milwaukee, WI, of nutrient composition 6-2-0 (N-P-K). Four hundred pounds were applied monthly by cyclone seeder on the 22nd of each month from May to September in both 1978 and 1979.

Specimens of sludge, soil, plants, and above-ground arthropods were collected monthly between the 18th and 21st of each month in 1979, starting immediately before the initial May sludge spreading and continuing through October. All samples were collected from three randomly selected locations within each plot and combined to make a monthly composite plot sample. Arthropods were collected by a vacuum collector process as described by BARRETT (1968) and were manually sorted from the litter. Plants were collected through the random location of a one-quarter square meter quadrat and the collection of all material within. Soil was collected by the removal of the upper 2 cm of soil from around the roots of the plants. All samples were preserved by freezing in glass containers until analysis.

Extraction was performed on 50 g aliquots of soil, 25 g aliquots of plants, and on the entire biomass of the arthropods collected. The aliquot was blended with a 1:1 hexane:acetone solution in a proportion of 2 mL solvent per gram of aliquot. After vacuum filtration, the soil and plant extracts were washed with 30 mL 2% saline and partitioned three times with 30 mL acetonitrile as described by THOMPSON (1977). Florisil and silicic acid fractionation were also done as described by Thompson. Fortified specimens of potting soil and greenhouse plants averaged 92% recovery from soil and 85% from plants.

To insure that arthropods were not contaminated by soil or detritus, individuals were cleaned and inspected manually under a 20 power microscope. Twelve of the arthropod samples were macerated and extracted as described by PETERSON et al. (1976). The remainder were suspended in 25 mL of the hexane-acetone solution and extracted in blenders before washing with saline and Florisil fractionation. Fortified specimens of a commercial hamburger-soybean mixture averaged 76% recovery of PCB.

Gas chromatography was performed using a tritium electron capture detector. Injector, column oven, and detector temperatures were all 200° C. All samples were injected into a 1/8 inch o.d., 3 m nickel column of 1.5% OV-17/1.95% OV-210 on 100-120 mesh Chromosorb W at a flow rate of 60 mL/min nitrogen. In addition, an auxiliary column of 5% OV-210 on Chromosorb W was used at 45-55 mL/min nitrogen for the separation and detection of PCBs. Retention times measured from chromatograms obtained on both columns were compared to Arochlor 1254 and 1260 standards obtained from the USEPA Environmental Toxicology Division, Research Triangle Park NC. Quantitation of the peaks was carried out on the OV-17/OV-210 column chromatograms.

Results were expressed numerically where they were above 20 ppb, while lesser detectable levels were called "trace" and were not used in the statistical analysis. Raw values were corrected for the percent recovery based on fortified specimens. The limit of detection was estimated to be approximately 8 ppb. The data were tested for significance using an analysis of variance (ANOVA) and the soil analyses were fitted to a line using least squares regression.

RESULTS

The Milorganite sludge was found to contain an average of 3.3 ppm (dry weight) PCB in the retention time range of peaks derived from a standard mixture of Aroclor 1254 and 1260. The range of values for six randomly chosen 50 lb bags was 3.1 to 3.4 ppm.

Soil samples collected from the treated plots showed trace amounts (i.e., less than 20 ppb) of PCB residues from the previous year's sludge treatment in the May 15, 1979, samples (Table 1). With additional sludge applications, the amount of detectable PCB in the soil increased to a quantifiable level in the July samples, and further to a mean of 34 ppb by October, three weeks after the final sludge application. From the control plots, one out of twelve samples showed trace residue levels.

Table 1. PCB residues in soil (ppb dry weight)

Treatment and Plot	May 15	June 15	July 19	Aug. 20	Sept. 18	Oct. 20
Control #1	N	N	N	N	N	N
Control #2	N	N	N	N	T	N
Sludge #1	T	20	21	22	36	39
Sludge #2	T	T	26	29	31	29
Sludge #3	T	T	19	27	29	35

"N" are no detectable PCB residues.
"T" or trace residues are detectable PCB residues, but at less than 20 ppb.

These soil concentrations are significant ($p < 0.05$) using ANOVA when compared to the controls. Least square regression of the PCB concentration vs. time in the treated soil samples yields an intercept of 14 ppb (with a 95% confidence interval of 6.4 to 21 ppb) as the May 22 PCB soil residue, or as the residue from the previous year's treatment. The slope of the regression is 0.14 ppb/day, or 4.0 ppb/treatment, and is significant ($p < 0.05$). The Pearson Correlation Coefficient between PCB concentration and time is 0.82.

Of the 72 plant samples collected, 67 were extracted and analyzed. Five received contamination in the laboratory during extraction and cleanup. The remaining 62 specimens had no detectable PCBs. In a separate experiment, three specimens of the Goldenrod, one from each treated plot, collected on September 18 were sorted and analyzed by leaves, stems, and flowers, and each subsample yielded no detectable PCBs.

For the arthropod analyses, none from the control plots showed detectable PCBs, nor did any animals collected in May and June. Among primary consumers, four samples from the treated plots from July and August showed a mean of 31 ppb PCB. In September, no residues were detected, and October arthropod samples were of insufficient biomass to be analyzed.

Arthropods in the May and June collections were primarily leafhoppers (Family Cicadellidae) and aphids (Family Aphidae) of the order Homoptera. In July and August, the greatest contribution to the biomass were crickets (Gryllus assimilus), Bean Beetles (Epilachna borealis), and grasshoppers (Dissostera carolina).

The secondary consumers were predominantly large spiders (Family Arachnida), Daddy Longlegs (Family Phalangida), Ladybugs (Adalia bipunctata), and Mantids (Mantis religiosa). Samples from the control plots yielded no detectable PCB residues, nor did samples from the treated plots in May, June, and September. Three samples from July and August did yield an average of 24 ppb. Differences between the primary and secondary consumers were not significant by ANOVA ($p < 0.10$).

DISCUSSION

The retention and increase of PCB residues in the soil confirms expectations that PCBs are relatively immobile and persistent in the A soil horizon. The lack of detectable PCBs above ground in the fescue grasses, Goldenrod, and miscellaneous plants confirms previous work with DDT. BROOKS (1974) found that DDT is taken up into those plant tissues with which it is in direct contact, in small amounts, but it is unlikely that the compound would be degraded or translocated unless the plant has a high oil or lipid content. This has been confirmed with PCBs in carrots (IWATA & GUNTHER 1976) and linseed (FINLAYSON & McCARTHY 1973; NASH 1974).

The largest contributors to the May and June insect biomass are aphids and leafhoppers, animals which rarely contact the ground and which showed no detectable PCBs. Whatever translocation of PCBs in plants may occur does not lead to biomagnification such that the limits of detection are reached. In July and August, the largest contributors are grasshoppers, bean beetles, and crickets, all larger animals that contact the ground frequently. The latter of these is a direct detritus consumer. The secondary consumers reflect uptake of the PCBs into these animals during these months. The bioconcentration values (1.3 times the soil for

primary consumers, 0.8 for secondary consumers for July and August) are considerably lower than those reported for aquatic insects (THOMPSON 1973) and for PCB-like compounds in beetles (GREICHUS et al. 1977).

Many factors are responsible for the movement of pesticides from soil to plants (NASH 1974). Other factors not presently considered, e.g., the route of application to both the ecosystem and to the organism (cutaneous contact vs. ingestion), climate, and life histories may also need to be included. One factor implied from this study is that PCBs can enter the food chain by direct consumption of the contaminated material (sludge or soil) rather than through transfer from plant to herbivore to carnivore.

Another point is that, although the insect residues are small, they can be significant when biomagnified. For example, an insect biomass of 1300 mg/m^2 at a PCB residue of 30 ppb means that there are 390 mg PCB/ha in the insects of a hypothetical field. If a half kilogram bird of prey consumes 10% of the field's insects in a season, then the bird accumulates a body burden of 39 mg, or a residue of 78 ppm. This does can be injurious to birds of prey and has been shown to cause eggshell thinning (MOORE 1974; HILL et al. 1976).

A review by TORREY (1979) recognizes this problem and proposes several criteria for the site selection and for the spreading procedures to be used for sludge disposal. These criteria include soil pH and composition, ground water and bedrock considerations, and drainage, moisture, and slope criteria which can reduce environmental hazards from the use of sludge as a fertilizer or soil conditioner.

Acknowledgements. The portion of this research conducted at the Miami University Ecology Research Center utilized equipment funded under U. S. EPA Grant No. R805445-01, awarded to C. S. Clark, Institute of Environmental Health, University of Cincinnati, and G. W. Barrett, Miami University, Principal Investigator.

REFERENCES

BARRETT, G. W.: Ecology 49:1019 (1968).

BROOKS, G. T.: Biological and Environmental Aspects. CRC Press, Inc., Cleveland, OH. (1974). p. 66-73.

FINLAYSON, D. G., and H. R. McCARTHY: Pesticide Residues in Plants. In Environmental Pollution by Pesticides (C. A. Edwards, ed.), Plenum Press, NY. (1973).

FOTH, H. D., and L. M. TURK: Fundamentals of Soil Science. J. Wiley and Sons, NY. (1972).

FURR, A. K., A. W. LAWRENCE, S. C. TONG, M. C. GANDOLOFO, R. A. HOFSTADER, C. A. BACHE, W. H. BUTENMANN, and D. J. LISK: Environ. Sci. Technol. 10:683 (1976).

GREICHUS, Y. A., A. GREICHUS, B. AMMAN, D. J. CALL, D. C. D. HAMMON, and R. M. POTT: Arch. Environ. Contam. Toxicol. 6:371 (1977).

HILL, E. F., J. D. WILLIAMS, and R. G. HEATH: Bull. Environ. Contam. Toxicol. 16:445 (1976).
IWATA, Y., and F. GUNTHER: Arch. Environ. Contam. Toxicol. 4:44 (1976).
MOORE, N. W.: Effects of Pesticides. In Conservation in Practice (A. Warren and F. B. Goldsmith, eds.), John Wiley & Sons, NY. (1974).
NASH, R. G.: "Plant Uptake of Insecticides, Fungicides, and Fumigants from Soils." In Pesticides in Soil and Water (W. D. Guenzi, ed.), Soil Sci. Soc. of America, Madison, WI. (1974).
PETERSON, J. E., K. M. STAHL, and D. L. MEEKER: Bull. Environ. Contam. Toxicol. 15:135 (1976).
THOMPSON, J. F.: Analysis of Pesticide Residues in Human and Environmental Samples. USEPA, Env. Toxicology Div., Research Triangle Park, NC. (1977).
TORREY, S.: Sludge Disposal by Landspreading Techniques. Noyes Data Corporation, Park Ridge, NJ. (1979).

Accepted September 8, 1981