Bull. Environ. Contam. Toxicol. (1986) 36:159–167

Bulletin of Environmental Contamination and Toxicology

Possible Sediment Scavenging of Chlordane and Contamination of the Aquatic Biota in Belmont Lake, New York

Lindsay W. Wood,[1] Peter A. Jones,[2] and Arthur Richards[1]

[1]Wadsworth Center for Laboratories and Research, New York State Department of Health, Albany, NY 12201 and [2]New York State Department of Environmental Conservation, 50 Wolf Road, Albany, NY 12233

The persistent organochlorine compound chlordane has been used extensively since its appearance on the market around 1947. On Long Island, New York, it is still used to control insects, particularly termites. Chlordane is poorly soluble in water. Estimates of the solubility range from 0.056 µg/L (Sanborn et al. 1976) to 9 µg/L (NRC 1982). The octanol/water partition coefficient (K_{ow}) for chlordane of 6.04 as determined by Sanborn et al. (1976) was identical to the value for its PCB analog (2,2', 3,3', 6-pentachlorobiphenyl) as reported by Rapaport and Eisenreich (1984). From the equations of Karickhoff et al. (1979), we estimated the logarithm of the organic carbon partition coefficient (K_{oc}) to be between 5.60 and 5.83 using a K_{oc} of 6.04 and a solubility of 9 µg/L. These properties suggest that chlordane readily adsorbs to clays and organic matter as do the PCBs (Di Toro and Horzempa 1982, Di Toro et al. 1983). Its affinity for organic matter in lake sediments can also be expected to be high.

In Belmont Lake, Long Island, New York, a shallow recreational lake, chlordane was reported to reach 5.2 µg/g in carp fillets and 0.38 µg/g in bass fillets (NYSDEC 1982) exceeding the applicable FDA guideline of 0.3 µg/g. As the use of this lake is presently impaired by excessive growths of aquatic weeds, restoration by dredging requires knowledge of the distribution, sources and fate of the chlordane.

MATERIALS AND METHODS

Belmont Lake is located in Suffolk County, Long Island, New York (Lat. 40° 43' 58'', Long. 73° 20' 32'') and is part of the south-draining Carll's River watershed. The lake is shallow (max. depth 1.3 m). Its surface area is 11.33 ha, its volume 77,000 m^3, its mean hydraulic retention time 2.0 to 2.9 days (FANS 1980). The retention time is highly variable with precipitation events, however. Surrounding soils are very permeable. The Upper Glacial Aquifer is located approximately 0.9 m below the lake surface at the outlet and is approximately 0.3 m above the surface near the inlet (Donaldson and Kosalka 1983).

The soils have a vertical permeability of 8.2 m/day, with a horizontal conductivity of 83.2 m/day (Tetratech 1977). As a result the lake is influenced heavily by groundwater which captures a lot of the 117 cm of annual precipitation. Groundwater may contribute as much as 70% of the total flow to the lake, and the drainage basin is reduced from its theoretical 73 km^2 to an effective size of 5 km^2. From July to September, the flow of the Carll's River to Belmont Lake is augmented by pumped groundwater which enters via a pipe in the river immediately above the lake. Pumping occurs at full force during the night and slows to a trickle during the day.

Since the 1930's when complete restructuring and cleaning of the lake occurred as a Public Works Project, sediments have accumulated over the hard sandy-gravel lake bottom to a depth of 60-100 cm in the north and 5-40 cm in the south. Total sediment volume is approximately 23,000 m^3 with a volatile solids content of from 11 to 30% depending upon location. Higher contents of volatile matter are found toward the north. Sediment texture in all areas is a sandy silt, with a slightly greater silt percentage in the south. Radioactivity dating of the sediments with ^{136}Cs and ^{210}Pb showed a discontinuity in the sediments deposited before 1954, possibly as a result of two hurricanes during 1954. The sedimentation pattern was normal after that date, with sedimentation rates of 0.19 $g/cm^2/yr$ (Jones and Wood 1983). The 60-cm deep sediment in several locations in the north was 5^{o} cooler at the bottom than at the top. The top had the same temperatures as the homothermous water column. The lower temperatures occurred in the lower 10 cm of the sedimentary column. No such differences were found in the southern areas (August 1981). This suggests groundwater infiltration in the northern areas and possible exfiltration in southern areas.

Dense growths of _Cabomba_ _caroliniana_ and _Potamogeton_ _bercholdii_ cover almost the entire surface of the lake from May through October. This growth reduces vertical mixing and stabilizes sediments during the growing season. _Potamogeton_ is predominant only in a restricted northern portion of the lake under the influence of the cooler groundwaters from flow augmentation.

The sampling grid for Belmont Lake is shown in Figure 1. The area of each grid unit is 929 m^2. Permanent stations were at locations H-6 and S-8. All other samples were identified by the grid coordinates. The variance inherent in the chlordane analyses was determined by a composite of 8 core samples from station H-6. The sediment was thoroughly mixed with a glass rod, allowed to settle, and the excess water decanted. Triplicate subsamples were analyzed and contained 0.49 ± 0.07 (actual values 0.42, 0.51 and 0.55) μg chlordane/g. The horizontal distribution of chlordane was determined from composites of 4 core samples at each of 6 stations, D/E-5, H/I-8, I/J-8, Q-10, T/U-9. The core contents were mixed, settled, and excess water decanted prior to placing a subsample into a hexane-washed jar with a Teflon lid. Vertical

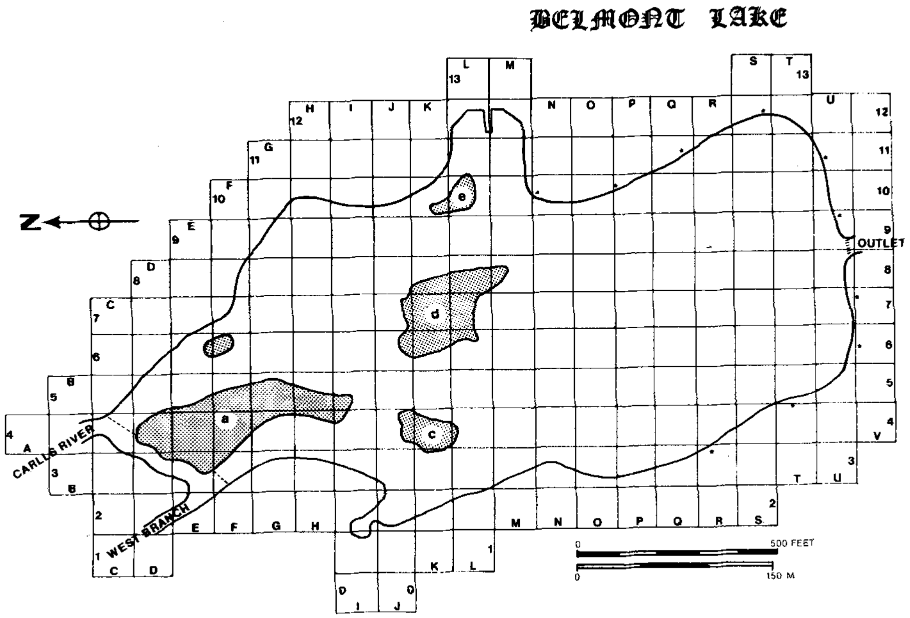

Figure 1. Map of Belmont Lake, Long Island, N.Y. showing the grid pattern referenced by an alpha numeric code for sample locations. Each grid square is 30.5 m on a side.

distribution was studied on one core each from stations H-6 and S-8 by sectioning into thirds. The sections from station H-6 were approximately 10 cm long and those from S-8 approximately 3 cm. Water samples were obtained from stations H-6, S-8, and from the Carll's River upstream and downstream of the lake. An additional sample was obtained from the flush pipe which had accumulated during the fall of 1981 and winter of 1982 was also collected and split into a top and bottom half.

Samples of soil from the parkland surrounding the lake were collected from lawn areas to the east and west of the lake as well as riverbank areas immediately upstream and downstream of the lake. These were collected in hexane-washed bottles by scooping along the surface of the soil in areas with little vegetative cover. Chlordane has not been used in the park since prior to 1971 (T. Lyons, NY State Department of Parks, Recreation and Historic Preservation, Albany, personal communication).

The water samples were extracted with 15% methylene chloride in hexane (EPA 1973; EPA-MSL 1978). Soil and sediment samples were prepared by the Veith and Kiwus (1977) modification of the Nielson-Kryger steam distillation technique. No further cleanup was required. Both techniques gave recoveries in excess of 85%. After concentration with a Kuderna-Danish apparatus the extracts were examined by gas chromatography-electron capture detection using a Hewlett-Packard model 5880 gas chromatograph with a 1.8 m column packed with 3% SE30 on (80-100 mesh) Gaschrom-Q (Alltech Associates Inc., Deerfield, IL).

RESULTS AND DISCUSSION

Chlordane levels in the superficial lake sediments decreased from 0.1 to 0.2 µg/g at the north end (H/I-8, D/E-5 and H-6) to 0.02 to 0.04 µg/g at the south end (I/J-6) (T-6, T/U-9, Q-10) with 0.06 µg/g in the central part (Table 1). The top and bottom sections of the sediment which had accumulated under the flush pipe contained 0.56 and 0.52 µg/g respectively. The sediments in the north and south ends of the lake showed a distinctly different vertical distribution of the chlordane. The top and middle thirds of the core from the northern station contained 0.18 and 0.11 µg/g respectively, whereas the bottom portion contained 0.58 µg/g. This portion of the core predates 1955. The core in the southern part had 0.05 µg/g in the upper and 0.04 and 0.03 µg/g respectively in the middle and lower thirds.

Soil concentrations of chlordane ranged from 0.003 µg/g along the river bank downstream of the lake to levels below the detection limit of 0.002 µg/g upstream of the lake (Table 2). Concentrations in the lawn and ballfield areas of the parkland to the east and west of the lake were also low.

All water samples showed chlordane to be below the detection limit of 0.11 µg/L (Table 2). This included the Upper Glacial Aquifer in this area.

Chlordane levels in the surface sediments of the north probably reflect the greater potential for accumulation in this area as opposed to the south. No evidence of scouring was observed in either the north or the south so that the deposits are stable. This may be largely due to lush growths of macrophytes which cover the deposit from shore to shore, producing a stabilizing effect on the sediments.

In the absence of measured concentrations in the water samples, a probable range of chlordane concentration in the aqueous phase can be estimated from the equation of Schwarzenbach and Westfall (1981):

$$\log K_p = 0.72 K_{ow} + \log f_{oc} + 0.49$$

where f_{oc} is the fraction of organic carbon in the sediment.

Table 1. Results of analyses for chlordane in lake sediments from Belmont Lake along with estimates of the corresponding aqueous concentrations corresponding to 11% and 30% carbon respectively (see text).

Location or Grid Coordinates	Sediment Conc. µg/g	Aqueous Conc. µg/L
Sediment composites		
Well Pipe (Top)	0.56	0.095 - 0.089
Well Pipe (Bottom)	0.52	0.088 - 0.082
T-6	0.02	0.003 - 0.003
T/U-9	0.04	0.007 - 0.007
Q-10	0.04	0.007 - 0.007
I/J-6	0.06	0.010 - 0.009
H/I-8	0.13	0.022 - 0.021
D/E-5	0.11	0.019 - 0.017
H-6	0.10	0.017 - 0.016
Sediment cores		
S-8 (Top)	0.05	0.009 - 0.008
S-8 (Middle)	0.04	0.007 - 0.006
S-8 (Bottom)	0.03	0.005 - 0.005
H-6 (Top)	0.18	0.031 - 0.028
H-6 (Middle)	0.11	0.019 - 0.017
H-6 (Bottom)	0.58	0.099 - 0.092

Table 2. Results from chlordane analyses on water and soils in the vicinity of Belmont Lake, N.Y. The grid coordinates refer to Figure 1.

Sample type (Conc. units)	Location or Grid Coordinates	Concentration
Water (µg/L)		
	Flush Pipe	LT 0.11
	River upstream	LT 0.11
	River downstream	LT 0.11
	River west branch	LT 0.11
	H-6	LT 0.11
	S-8	LT 0.11
Soils (µg/gm)		
	West of Lake (Lawn)	LT 0.002
	East of Lake (Field)	0.002
	East of Lake (Lawn)	0.003
	Riverbank (Upstream)	LT 0.002
	Riverbank (Downstream)	0.003

If volatile solids content is used as an estimate of organic carbon, K_p values of 5.88 to 6.32 are obtained for volatile solid contents between 11 and 30% and a K_{ow} of 6.04. The equation:

$$c = s/K_p$$

(Schwarzenbach and Westfall 1981) was then used to estimate maximum concentrations in the water (c) from the concentration in the sediment (s). Over estimation of the organic carbon can result from the assumption that its content equals that of volatile solids.

If the decrease of chlordane concentrations from the bottom to the top of the sediments in the northern part of the lake is taken as an indication of chlordane additions from groundwater as suggested by the temperature gradient in these sediments, then the aqueous chlordane concentrations from point of upwelling to the deposit to point of exfiltration from the deposit in the north would decrease from a calculated range of 0.092 - 0.099 to a range of 0.028 - 0.031 µg/L. In the south, the remaining estimated concentration range of 0.008 - 0.009 µg/L could decrease to 0.005 during descent through the deposit to the groundwater table. This lack of a similar sharp increase in chlordane levels at the bottom of southern deposits also suggests that the chlordane did not enter the lake during the initial accumulation of the sediment deposit. Indeed, the concentration in this area actually decreases with sediment depth.

The data suggest the existence of a source of chlordane north of the lake, and the source is still present as the substance is still being injected (to be terminated shortly) into the ground for termite control bypassing the topsoil (L. Skinner, NYSDEC. News Release of 7 September 1984, Personal Communication). The low concentrations in the surface and riverbank soils further point to surface runoff as not being a significant source. One may therefore hypothesize that chlordane in ground water passing from the sand-gravel substratum into the relatively organic-rich sediments is the probable source. The organic material under the flush pipe would similarly be contaminated largely from groundwater via seepage from the river bed, which forms 70% of the flow of the Carll's River. The sediment under the flush pipe would be periodically flushed during the summer to be broadcast upon the surface of the deposit. This broadcast and deposition of recent sediments along with scavenging from the water by the deposit and suspended particles would explain the observed horizontal gradient. Exfiltration near the lake outlet would lead to greater scavenging of chlordane at the surface of the sediments there.

If scavenging of chlordane from dilute concentrations in groundwater serves as a first concentration step, then this represents a physico-chemical magnification of this substance. It has been shown that heat-killed algal cells resemble living cells in their ability to scavenge chlordane from water (Glooschenko et al. 1979). Biomagnification could then proceed from this initial step via benthic organisms. In many sediment cores fresh burrows made by aquatic oligochaetes (''sludgeworms'') were found at all depths to the bottom of the sediments (35 cm or more). These observations were made using clear plastic core tubes so that burrow formation and distribution could be observed.

These oligochaetes select organic particles and associated bacteria for food and energy (Brinkhurst and Chua 1969, Brinkhurst et al. 1972). Recent work has pointed to high concentration by these worms of substances with K_{ow} values around 6 (Oliver 1984). Other organisms have similar high concentrating abilities for high-K_{ow} compounds modified somewhat by a food web position factor (Fox et al. 1983). These worms which occur at population densities of 13 to 436 individuals/m^2 and other forms (insect larvae, mollusks, etc.) are more favored by benthic feeding fish such as carp than bass. In 5 samples from the northern portion of Belmont Lake, the number of macrobenthos plus macroinvertebrates associated with macrophytes ranged from 18 - 688 individuals/m^2. In the southern portion, with more of the invertebrates associated with the plants, the range was 56 - 482 (Jones and Wood 1983). The biomagnification potential for chlordane would be greater in northern areas (0.1 - 0.2 µg/g) than in southern areas (0.02 - 0.05 µg/g). Consequently, benthos-feeding fish (such as carp) through resource partitioning would tend to harvest higher chlordane levels than species (such as bass) which tend to feed more in the water column. Both contact and ingestion have been identified as mechanisms for bioaccumulation of PCBs and 2,4,-D (Haque and Ebling 1983). Intake and excretion are also related to observed contamination levels in biota and appear to be species specific. At a given concentration, algae, zooplankton, and fish each accumulated chlordane to different levels (Moore et al. 1977). For goldfish, elimination of chlordane was through biliary pathways and through the gills, kidney and feces. In Belmont Lake, the concentrations in the fish probably result from both food and contact uptake leading to a steady state with excretory losses.

Our hypothesis suggests that both physico-chemical and biomagnification mechanisms in Belmont Lake are responsible for the significant contamination of local fish (NYSDEC 1982). Scavenging of chlordane from groundwater inflows by sorptive partitioning to organic matter in the sediments may be the first step. This concentrate then would become available for biomagnification through the food web to fish.

Acknowledgments. Mr. W. Ahearn was very helpful in the collection and processing of samples in the field. T. Lyons, G-Y. Rhee, G. W. Fuhs provided helpful comments on the manuscript. The work was supported in part by a Clean Lakes Grant from EPA (S 002260010).

REFERENCES

Brinkhurst RO, Chua KE (1969) Preliminary investigation of exploitation of some potential nutritional sources by three sympatric tubificid oligochaetes. J Fish Res Bd Canada 26:2659-2668

Brinkhurst RO, Chua KE, Kaushick NK (1972) Interspecific interactions and selective feeding by tubificid oligochaetes. Limnol Oceanogr 17:122-133

Di Toro DM, Horzempa LM (1982) Reversible and resistant components of PCB adsorption-desorption isotherms. Environ Sci Technol 16:594-602

Di Toro DM, Horzempa LM, Casey ML (1983) Adsorption and desorption of hexachlorobiphenyl. EPA-600/S3-83-088, Environmental Research Laboratory, USEPA, Duluth, Minnesota (Project summary)

Donaldson CD, Kosalka EJ (1983) Open File Report 82-163, Plate 1A west, US Department of the Interior, US Geological Survey, Syosset, New York

Environmental Protection Agency (1973) Method for organochlorine pesticides in industrial effluents. MDQUARL U.S.E.P.A., Cincinnati, Ohio

Environmental Protection Agency - EMSL (1978) Methods for benzidine, chlorinated organic compounds, pentachlorophenol and pesticides in water and wastewater. Interim, pending issuance of methods for organic analysis of water and wastewater. EPA-EMSL, Cincinnati, Ohio

Fox, ME, Carey JH, Oliver BG (1983) Compartmental distribution of organochlorine contaminants in the Niagara River and the western basin Lake Ontario. J Great Lakes Res 9:287-294

FANS (1980) Study to determine the necessity, and if applicable, the methods of mitigating the decrease in streamflow and related effects associated with sewering Suffolk County, Long Island, New York. Suffolk County Dept Environ Contr, Melville, New York

Glooschenko V, Holdrinet M, Lott JNA, Frank R (1979) Bioconcentration of chlordane by green alga <u>Scenedesmus quadricauda</u> Bull Environ Contam Toxicol 21:515-520

Haque A, Ebling W (1983) Uptake, accumulation, and elimination of HCB and 2,4-D by the terrestrial slug, <u>Deroceras reticulatum</u> (Muller) Bull Environ Contam Toxicol 31:727-733

Jones PA, Wood LW (1983) A diagnostic-feasibility study of Belmont Lake, Suffolk County (Long Island), New York with a view towards its restoration. pp 36-156. In Diagnostic Feasibility Study Report and Draft Environmental Impact Statement (DEIS) on the proposal to restore Belmont Lake in Belmont Lake State Park, Town of Babylon, NYS Office of Parks Recreation and Historic Preservation, Albany, New York

Karickhoff SW, Brown DS, Scott TA (1979) Sorption of hydrophobic pollutants on natural sediments. Water Res. 13:241-248

Moore R, Toro E, Stanton M, Khan MAQ (1977) Absorption and elimination of ^{14}C-alpha- and gamma-chlordane by a freshwater alga, daphnid, and goldfish. Arch Environ Contam Toxicol 6:411-420

National Research Council (1982) Committee on Toxicology, Board of Toxicology and Environmental Health Hazards, Commission on Life Sciences, National Research Council, Washington, D.C. 75 pp

NYSDEC. Toxic substances in fish and wildlife (1982) (Nov. 1, 1981 - Apr. 30, 1982) New York State Department of Environmental Conservation report 82-2 (DEP) 5:17

Oliver BG (1984) Uptake of chlorinated organics from anthropotenically contaminated sediments by oligochaete worms. Can J Fish Aquat Sci 41:878-883

Rapaport RA, Eisenreich SJ (1984) Chromatographic determination of octanol-water partition coefficients (Klow's) for 58 polychlorinated biphenyl congeners. Environ Sci Technol 18:163-170

Sanborn, JR, Metcalf RL, Bruce WN, Lu P-Y (1976) The fate of chlordane and toxaphene in a terrestrial-aquatic model ecosystem. Environ Entomol 5:533-538

Schwarzenbach RP, Westall J (1981) Transport of nonpolar compounds from surface water to groundwater. Laboratory sorption studies. Environ Sci Technol 15:1360-1367

Tetratech, Inc. (1977) Stream aquifer model of Carll's River basin, Long Island, New York Rept No TC-686, Lafayette, California

Veith GD, Kiwus LM (1977) An exhaustive steam-distillation and solvent-extraction unit for pesticides and industrial chemicals. Bull Environ Contam Toxicol 17:631-636

Received February 5, 1985; accepted February 25, 1985.