Bull. Environ. Contam. Toxicol. (1992) 49:217–223

Environmental Contamination and Toxicology

Chlorinated Pesticide Residue Occurrence and Distribution in Mosquito Control Impoundments along the Florida Indian River Lagoon

Tsen C. Wang,[1] Marisa E. Hoffman,[1] Jim David,[2] and Randy Parkinson[3]

[1]Harbor Branch Oceanographic Institution, Fort Pierce, Florida 34946, USA; [2]St. Lucie County Mosquito Control District, Fort Pierce, Florida 34982, USA, and [3]Florida Institute of Technology, Melbourne, Florida 32901, USA

The advent of DDT as a contact insecticide in the early 1940's marked the beginning of the era of synthetic organic pesticides. A large family of organochlorine pesticides emerged and were widely used. These pesticides included DDT, lindane, heptachlor, heptachlor epoxide, dieldrin and aldrin. The migration of these pesticides into various areas of the environment has generated considerable public apprehension concerning their fate and effect. Certain pesticides applied to crops and soil for pest and mosquito control do not remain on site, but are transported in runoff water and on eroded soil particles into receiving waters (Grzenda et al 1965, 1966; Lauer et al 1966; Nichloson et al 1966, 1967), where they may be absorbed directly by living organisms (Kolipinski et al 1971; Seba et al 1969; Wang et al 1983), that are subsequently consumed by larger organisms. In this manner, organochlorine pesticides may be passed onto secondary consumers and up through the food chain. Several instances of poisoning have been reported as a final step in the biological magnification process.

Due to the persistent and long residual effectiveness of the pesticides, quantities of DDT, lindane, dieldrin, and chlordane mixed with fuel oil were applied from both ground (DDT) and air (DDT, lindane and dieldrin) by the St. Lucie County Mosquito Control District in the late 1940's and early 1950's. Unknown quantities were also applied to the marshes by the military during World War II. Of concern in this study are the concentrations of these pesticides which may still remain within the marsh substrate, and the mobilization of these residues.

The goal of this project was to: (1) document the presence or absence of chlorinated pesticide residues within the marsh substrate of 18 St. Lucie County mosquito control impoundments along the Indian River Lagoon (IRL), (Figure 1); (2) document the mobility of these pesticides into the IRL under contrasting marsh management techniques (well-flushed vs. poorly flushed), and (3) document the effect of standard impoundment management techniques on Indian River water quality.

Send reprint requests to T. Wang at the above address.

MATERIALS AND METHODS

Two random sampling sites for each impoundment were selected for sampling both core sediment and core water samples. Each core sample was approximately 6 ft in depth and was subdivided into three sections (top, middle, and bottom) for analysis. During the screening phase, a total of 34 water samples and 92 sediment samples were analyzed to screen for the presence and distribution of pesticides in 18 impoundments.

Two impoundments were selected for detailed study. One impoundment (#1) covers 639 acres and is connected to the IRL by five culverts (Figures 1 and 2). It is considered to be a poorly-flushed impoundment (acreage:culvert = 128) since the impounded water levels oscillate only a few centimeters over the tidal cycle and do not replicate the estuarine tidal range (IRL tidal range 1.3-1.5 ft). In contrast, impoundment #2 covers 188 acres and is connected to the IRL by seven culverts (Figures 1 and 2). It is considered to be a well-flushed impoundment (acreage:culvert = 27) with little tidal dampening (<10%) and inconsequential time lag observed.

In addition to the detailed sampling of the marsh substrate, three field sessions focused on the export and import characteristics of the exchanging waters. The two impoundments selected for detailed study (imp. #1 and #2), and the IRL estuary, were sampled in the viscinity of the exchange-point-culverts. Water samples were collected inside the impoundments at the marsh edge (50 feet from the culverts), at the culvert mouth, and 100 feet offshore in the IRL estuary. Collections were made: (1) at the end of the open season, when the marsh exchanges freely with the estuary through open 76 cm diameter culverts; (2) at the end of the closed season, when the marsh is initially opened after several months of relative isolation from the estuary, and (3) at mid-season drawdown, when the impoundment(s) are completely drained during the mid-closure period (temporally between items 1 and 2 above and performed for impoundment #2 only, in this study).

Water samples were centrifuged at 8000 G for 25 min at 10°C to separate suspended solids from the water. A 900 ml water sample was extracted with 60 ml of methylene chloride three times. The extract was then passed through a Na_2SO_4 and florisil column for clean-up procedures. Activated copper was added to remove sulfur compounds prior to gas chromatographic analysis (Mills et al 1972, Law et al 1979, USEPA 1971). The minimum detectable concentration for each pesticide compound is 0.01 µg/l. The mean recovery and relative standard deviation for each compound ranged between 84% to 99% and 7% to 16%, respectively.

EPA method 3540 (USEPA 1986) was used to extract sediment samples for chlorinated pesticide analysis. A dried sediment sample (100 g) was soxhlet extracted with 300 ml hexane/acetone (1:1) mixture for 24 hours. The extract was then passed through Na_2SO_4 and florisil columns. Copper was also added to remove sulfur. A 30 m X 0.25 mm DB-5 fused silica column equipped with an electron capture detector

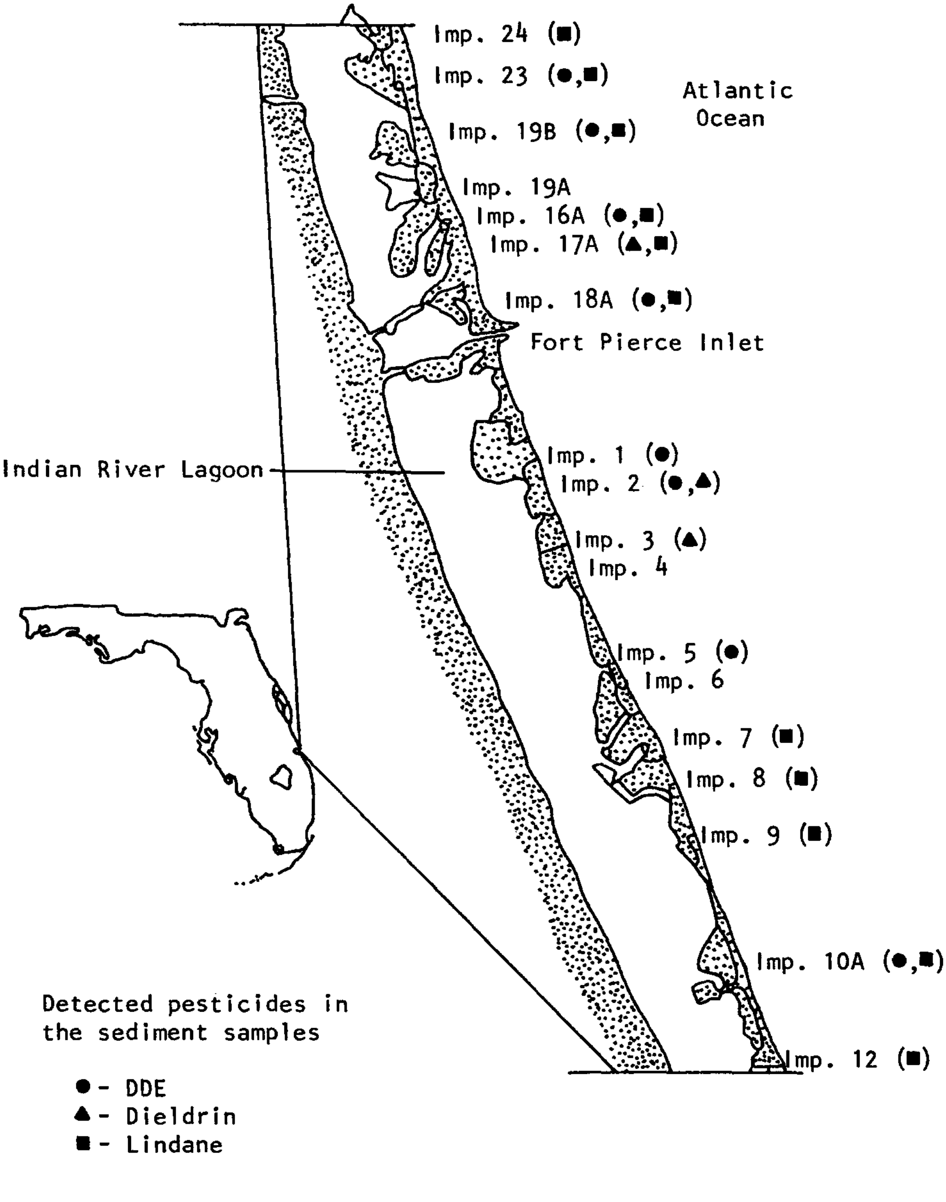

Figure 1. Location and distribution of chlorinated pesticides in each impoundment

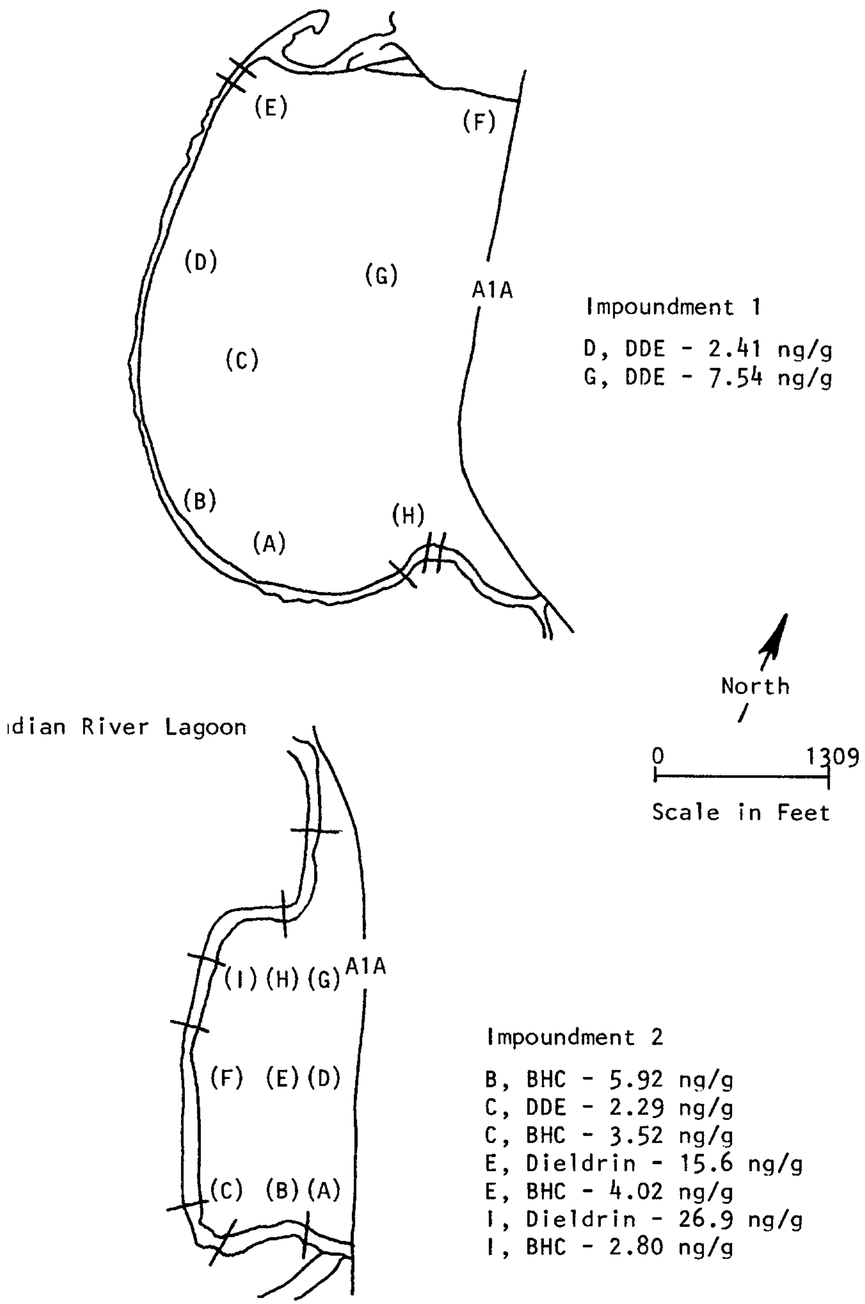

Figure 2. Transect sampling sites for impoundments 1 and 2

in a Perkin Elmer Sigma 3B gas chromatograph was used to analyze the samples. Argon/methane (6 ml/min) was used as carrier gas. The temperature program was set at 180 °C for 6 min. to 280 °C at a rate of 3 °C/min. A gas chromatograph/mass spectrometer (A DB-5 capillary column equipped with Varian GC 3400 and Finnigan ITDS 806A) was used to confirm sample analysis. The minimum detectable concentration for sediment samples ranged between 0.17 ng/g for α-BHC to 0.49 ng/g for endrin. The mean recovery and relative standard deviation of sediment analysis for chlorinated pesticides ranged between 77% to 98% and 6% to 13%, respectively. The analytical standard for each compound was obtained from Supelco, Inc. For each set of samples, a method blank, a spiked sample and the unknown samples were performed. Prior to each unknown sample analysis, dibutyl chlorendate was spiked as a surrogate standard to monitor sample extraction efficiency.

RESULTS AND DISCUSSION

A total of ten sediment samples from eight impoundments were found to contain DDE. The concentration ranged between 31.2 ng/g at impoundment 23 (site 1, top layer, impoundment open year-round) to 1.87 ng/g at impoundment 10A (site 2, top layer, impoundment open year-round). Figure 1 shows that the DDE occurred mostly in the sediments of the northern mosquito impoundments, which is adjacent to the military training areas used in World War II. Impoundments #1, #2, #5, #10A, #16A, #18A, #19B, and #23 were detected with DDE at least once during the study period.

A total of six sediment samples from three impoundments (#2, #3, #17A) contained dieldrin. Figure 1 shows dieldrin contamination mostly occurred in the middle section of the study area, in which an aerial application of deildrin pellets was performed for a sandfly control experiment in the mid 1950's. Top layer sediment samples of both impoundments #2 and #3 had a dieldrin concentration of 34.0 and 43.2 ng/g, respectively.

Lindane was also detected in the sediment. A total of 33 sediment samples from 11 impoundments were found to contain lindane. The compound was distributed mostly at the southern and northern ends of the study area (impoundments #7, #8, #9, #10A, #12, #16A, #17A, #18A, #19B, #23 and #24) as shown in Figure 1. Lindane was used as a general aerial larvicide over most of the barrier island marshes by Mosquito Control, from the latter-1950's to the mid-1960's. The concentration was generally higher in the top layer samples and gradually decreased in the bottom layer samples. The highest detected concentration of lindane was 34.4 ng/g at impoundment 12 (site 2, top layer), declining to 9.4 ng/g in the bottom layer at the same site.

Interstitial water samples were collected at each core site (except #18A which was collected adjacent to the site). None of the interstitial water samples contained detectable levels of pesticide residues.

Impoundments #1 and #2 were selected for further detailed analysis to determine the extent, areal distribution and mobility of pesticide residues. Figure 2 depicts the transects for the sampling sites for impoundments #1 and #2. Eight and nine sampling sites were selected in impoundments #1 and #2, respectively (approximately 456m and 183m apart). Top layer sediment and core hole water samples from each site were collected. A total of 17 water and 17 sediment samples each were analyzed. DDE was detected in the sediments of two sites from the northern end of impoundment #1, an area adjacent to early development, which may have had ground adulticiding with DDT performed. The concentration was 2.41 and 7.54 ng/g at transect site D and G, respectively. Both of these samples, plus the sample from the initial system-wide random sampling site, indicate that DDE contamination mostly occurred at the north end of impoundment #1. Lindane was found in 4 samples (B, C, E, and I) collected at impoundment 2. The concentration ranged from 2.80 ng/g at transect I to 5.92 ng/g at transect B. Dieldrin was found in two samples from impoundment #2. A concentration of 15.6 ng/g and 26.9 ng/g was detected at transect E and transect I, respectively. DDE (2.29 ng/g) was also found at transect C, impoundment 2.

Water exchanging between the impoundments and the estuary was also examined, in order to determine the extent of pesticide export (mobilization) and the degree to which such transport might be linked to water-borne organic export. Water samples in impoundments #1 and #2 were collected: (1) at the end of the open session (March 8, 1990 for impoundment 1 and April 17, 1990 for impoundment 2); (2) at the Mid-closure period during a drawdown of impoundment waters (impoundment #2 only, on July 12, 1990); (3) at the end of the closure period during initial reopening of the impoundment (on October 8, 1990 for both impoundments). Two culverts were selected for analysis from each impoundment. A total of 88 water samples were analyzed for pesticides. The pesticide concentration in all of 88 water samples analyzed was non-detectable.

Chemical analysis of 53 sediment cores taken from 18 St. Lucie County mosquito control impoundments suggests organochlorine pesticide distribution is widespread. Eighty-nine percent of the impoundments contained pesticides. However, the system-wide distribution is very scattered and and the pesticides do not appear to be mobilized in any detectable manner. Detailed analysis of impoundments #1 and #2 suggest the distribution of pesticide residue within each impoundment is also variable. Thirty-three percent of the core samples in impoundment 1 and 60% of the core samples in impoundment 2 contained pesticide residue. Geographically, lindane appears to be concentrated in the northern and southern areas, while dieldrin (a residual of sandfly control experiments conducted in the mid 1950's) is more prevalent in the central areas.

In most cases the concentration of organochlorine compounds in the sediment, if present, decreases with depth. Pesticides were only found in subsurface samples if the overlying surface sample contained pesticides. The project also concludes that water management of the impoundments does not appear to have any affect of pesticide mobility from the sediments. Estuarine water circulating through the impoundments does not contain detectable levels of pesticides, thus this study provides no evidence that the multi-million-dollar restoration programs ongoing and/or planned for the Indian River Lagoon impoundments (whose purpose is to restore tidal flushing to the impounded wetlands to improve water quality and habitat value), could result in pesticide mobilization and export to the estuarine system through water-borne transport mechanisms.

Acknowledgments. The project was supported by the Florida Costal Zone Management and the St. Lucie County Mosquito Control District. This paper is Harbor Branch contribution No. XXX.

REFERENCES

Grzenda AR, and Nicholson HP (1965) The distribution and magnitude ofinsecticide residues among various components of a stream system. Proc Water Resour Pollut Contr Conf 14:165-174

Grzenda AR (1966) A five-year statistical survey of cotton insecticides usage in a large Alabama watershed. J Georgia Entomol Soc 1:1-11

Lauer GJ, Nicholson HP, Cox WS, Teasley JI (1966) Pesticide contamination of surface water by sugar cane farming in Louisiana. Trans Amer Fish Soc 95:310-316

Law LM, Goerlitz DF (1979) Microcolumn chromatographic cleanup for the analysis of pesticides in water. J of the AOAC 53(6):1276-1281

Mills PA, Bong BA, Kanps LR, Burke JA (1972) Elution solvent system for Florisil column cleanup in organochlorine pesticide residue analysis. J of the AOAC 55(1):39-43

Nichloson HP, Webb HJ, Lauer GJ, O'Brien RE, Grzenda AR, Shanklin DW (1966) Insecticide contamination in a farm pond. Part I. Origin and duration. Trans Amer Fish Soc 91:213-222

Nichloson HP, Grzenda AR, Lauer GJ, Cox WS, Teasley JI (1967) Water pollution by insecticides in an agricultural river basin. I. Occurrence in river and treated municipal water. Limnol Oceanogr 9:310-317

Kolipinski MC, Hinger AL, Yates ML (1971) Organochlorine insecticide residues in Everglades National Park and Loxahatchee National Wildlife Refuge, Florida. Pesticide Monitoring Journal 5(3):281-288

Seba DB, Corcoran EF (1969) Surface slicks as concentrations of pesticides in the marine environment. Pesticide Monitoring Journal 3(3):190-193

Wang TC (1983) Toxic substance monitoring in the Indian River Lagoon, Florida. Florida Scientist 46(3):286-295

Wang TC, Johnson RS, Krivan JD (1983) Residues of polychlorinated biphenyls and DDT residues in water and sediment of the St. Lucie Estuary, Florida. Journal of Pesticide Monitoring 13(2):69-71

US Environmental Protection Agency (1986) Test methods for evaluating solid waste, Method 3540. Lab Manual Vol 1B SW-846

US Environmental Protection Agency (1971) Methods for organic pesticides in water and wastewater. National Environ Research Center. Cincinnati, OH

Received November 30, 1991; accepted January 5, 1992.