SOURCES AND FATE OF ORGANOCHLORINE PESTICIDES IN THE GREAT LAKES: AN OVERVIEW

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Introduction

Organochlorine (OC) pesticides were widely used in the Great Lakes Basin in the past. Monitoring of fish, bird, and mammalian species in the Great Lakes during the 1970s provided the first alert of adverse health effects due to exposure to OC pesticides such as DDT and dieldrin. Due to the persistence, toxicity, bioaccumulative properties, and long-range atmospheric transport of these pesticides, their production and use in North America were largely banned in the late 1970s. However, residues of the OC pesticides continue to be detected in most environmental matrices in the Great Lakes. The unintended effects of OC pesticides continue to be of concern, and evaluation of the risk to humans and to the environment from current levels of exposure remains the subject of continued scientific research in the Great Lakes Basin. Here we summarize trends in OC pesticide use, sources, environmental levels and loadings in the Great Lakes Basin.

Results and Discussion

Trends in the Use of OC Pesticides

Organochlorine insecticides were the major class of compounds used in agriculture in the USA and Canada during the 1950s and the 1960s. DDT was the predominantly used insecticide in agriculture in the USA and Canada until the early 1970s, when most uses of DDT were banned in Canada and the USA. The use of aldrin, dieldrin, mirex, heptachlor and heptachlor epoxide were banned in the late 1970s. Although the domestic use of chlordane was curtailed in 1988, chlordane continued to be produced in the USA for export until 1997. Toxaphene was prohibited from use in 1983 in the USA, and it was de-registered in 1982 in Canada.

The past usage of OC insecticides was large enough to cause widespread environmental contamination. Whereas environmental concentrations of OC pesticides in the Great Lakes Basin have generally declined during the past 20 years, and the current concentrations in surface water are below the drinking water standards, concerns nevertheless remain because these substances persist in the environment and accumulate in the food chain. OC insecticides can magnify by a factor of several tens to hundreds in predatory organisms relative to their prey. There continue to be fish consumption advisories based on unacceptable levels of OC pesticides in sport and commercial fish from the Great Lakes.

Atmospheric transport from agricultural regions in the USA and Canada, where these pesticides were used extensively in the past, continues to be a source in the Great Lakes.[1] Moreover, a few former production sites of OC pesticides are located in the Great Lakes Basin. There are 20 aldrin-, 29 dieldrin-, 23 chlordane-, 44 DDT-, and 3 mirex-contaminated hazardous waste sites located in the Great Lakes Basin. Hazardous waste sites with elevated levels of these pesticides continue to represent sources to the Great Lakes, either through leaching into groundwater, soil runoff, and erosion, or atmospheric transport. A few OC insecticides that are still in use in the Great Lakes region are-methoxychlor, dicofol, endosulfan, and lindane (γ-HCH).

Environmental Levels and Trends of OC Pesticides.

There is a considerable body of literature documenting the accumulation of OC pesticides in the Great Lakes since the late 1960s. Currently, several international, national, regional, and local monitoring programs are in place in the Great Lakes region, to monitor sources, trends, distribution, fate, and effects of OC compounds. The USEPA's Great Lakes National Program Office and the USGS's Great Lakes Science Center have been conducting joint monitoring programs since 1977 using fish, particularly lake trout and walleye as biomonitors. Results of fish biomonitoring studies from 1977 to 2003 have been published.[2,3] DDT is the major pesticide found in fish and birds in the Great Lakes. On a by-lake basis, lake trout from Lake Michigan contain the highest concentrations of DDT (Fig. 1) [3], followed by those from Lake Ontario. Similarly, Environment Canada and the Canadian Wildlife Service have been

conducting biomonitoring programs, to detect the trends of persistent organic pollutants in fish and in herring gull eggs, since the 1970s.[4] Trend studies have shown that most of the OC pesticides in salmonid fishes declined rapidly until the 1980s, and they then leveled off in the 1990s.[5] Similarly, double crested cormorant eggs collected from Lakes Ontario, Erie, and Superior showed a decline in DDT concentrations from 1970 to 1989, but levels thereafter remained stable.[6] The data also indicate that the rate of this decline has now slowed. While the trends of contamination levels for most OC pesticides follow a predictable trend of highest contamination in Lake Michigan followed by Lake Ontario, toxaphene levels are historically the highest in Lake Superior. No significant decline in toxaphene concentrations was found in fish from Lake Superior during 1982-1992, whereas concentrations in fish from the other Great Lakes declined after the ban on the use of toxaphene in the USA in 1982.[7]

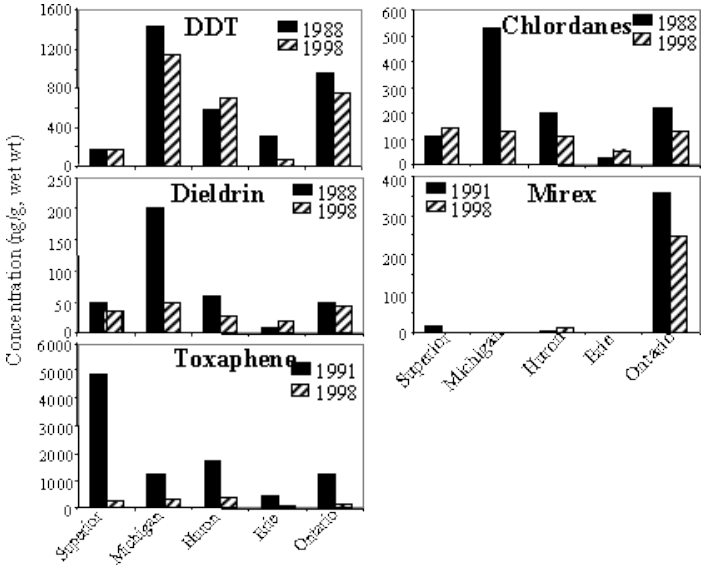

Fig. 1. Concentrations of organochlorine pesticides in lake trout and walleye (Lake Erie only) collected in 1988 and 1998. For mirex and toxaphene, 1991 concentrations are presented instead of 1988 values.[2,3]

Trends in Environmental Loadings

Since the cessation of production and use of OC pesticides, direct discharges to the Great Lakes have greatly diminished. The relative steady state in contaminant levels in the 1990s indicates that these chemicals are still being released and/or recycled through the Great Lakes ecosystem. Atmospheric deposition, agricultural runoff, the slow movement (leaching) of discarded stocks of pesticides from landfill sites into the Great Lakes continue to be major indirect sources of contamination. To investigate atmospheric deposition, the USEPA and Environment Canada operate a monitoring network that measures levels of toxic substances in the air of the Great Lakes region. The Integrated Atmospheric Deposition Network (IADN) was created under Annex 15 of the Great Lakes Water Quality Agreement. Samples of air and precipitation have been taken since 1990, with a primary goal of measuring several persistent organic pollutants, including 22 chlorinated pesticides.[1] IADN publishes a loadings report every two years, summarizing these measurements and providing estimates for the atmospheric deposition of selected pesticides (α- and γ-HCHs, α- and γ-chlordanes, dieldrin, *p,p'*-DDE, *p,p'*-DDD, and *p,p'*-DDT) in addition to several other compounds to each of the Great Lakes. On the basis of this network and individual process studies, there is substantial evidence to support the idea that atmospheric transport and deposition of pesticides contribute significantly to, and in some cases are the dominant source of, contaminant inputs in surface waters of the Great Lakes.[8] The recent IADN report covers data through 1998,[8] which for the first time have been expressed on a Great Lakes Basin-wide basis, through summation of the total deposition of each pesticide measured over the five Great Lakes (in kg/yr) for the period 1992 to 1998 (Table 1). Total deposition of a-HCH showed a trend of decrease, from 950 kg/yr in 1992 to 210 kg/yr in 1998. Dieldrin and DDT had negative total deposition across time, indicating that the lakes are acting as a source of these chemicals to the atmosphere.

Table 1. Total deposition (kg/yr) of selected organochlorine pesticides in the Great Lakes in 1998 .

Pesticide	Superior	Michigan	Huron	Erie	Ontario	All Lakes
DDT (DDD+DDE+DDT)	11.8	18.9	14.7	-36	14.8	-5.4
Dieldrin	-250	-	-	-70	-120	-440
γ-HCH	7.6	200	170	39	36	453
α-HCH	-460	220	-42	95	-23	-210

Negative values indicate loss or volatilization.[9]

Among the pesticides studied, lindane showed the highest downward flux, which is explained by current use of this pesticide in agriculture.[10] Overall, the IADN data indicate that the net loadings of most banned OC pesticides to the Great Lakes are decreasing, and are in some cases approaching zero. In many cases, the dominant factors of gas absorption and volatilization are near steady-state, with the amount going into the lakes equaling the amount coming out.[11] When a loading nears zero, this does not mean that pollutant levels in the lakes are at zero. The pollutants are still present, cycling continuously and evenly between the air and water. In the absence of new sources to the Lakes, concentrations in both air and water, and therefore also in wildlife, are expected to continue to decline, likely at rates determined by the decline of global air concentrations, which in turn is governed by the temperature-mediated volatilization of residues from soil reservoirs and the processes of destruction and long-term burial into the earth.

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