Pesticides in the Nation's Rivers, 1975–1980, and Implications for Future Monitoring

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Pesticides in the Nation's Rivers, 1975–1980, and Implications for Future Monitoring

By Robert J. Gilliom, Richard B. Alexander, *and* Richard A. Smith

Abstract

Water samples were taken four times per year and bed-sediment samples two times per year during 1975–80 at 160 to 180 stations on major rivers of the United States. Samples were analyzed for 18 insecticides and 4 herbicides, which together accounted for about one-third of the total amount of all pecticides applied to major crops during 1975–80. Fewer than 10 percent of almost 3,000 water samples and fewer than 20 percent of almost 1,000 bed-sediment samples contained reportable concentrations of any of the compounds. The patterns of detection result from a combination of widely variable detection capabilities, chemical properties, and use. Most detections in water samples were of relatively persistent yet soluble compounds: atrazine (4.8 percent of samples), diazinon (1.2), and lindane (1.1). Most detections in bed-sediment samples were of the hydrophobic and persistent insecticides: DDE (17 percent of samples), DDD (12), dieldrin (12), chlordane (9.9), and DDT (8.5). Only for atrazine in water, and for DDE, DDD, DDT, and chlordane in bed sediments, were geographic patterns of detection correlated ($p \leq 0.10$) with use on farms. Detections of organochlorine insecticides in both water and bed sediments appear to have erratically but gradually decreased during 1975–80. For the 1975–79 period, more stations had downtrends than had uptrends in bed-sediment levels of organochlorines. No clear trends were evident in concentrations of organophosphate insecticides or herbicides in either water or bed sediments. Findings suggest that future pesticide monitoring efforts must be responsive to changes in pesticides used and to geographic patterns of use. Different types of monitoring approaches are necessary for chemicals having different chemical and physical properties. Before an effective dynamic monitoring effort can be designed, however, selected case studies are needed to characterize and refine sampling and analytical capabilities for different types of chemicals, river environments, and sample types.

INTRODUCTION

A national network for the monitoring of pesticide residues in river waters and bed sediments, the Pesticide Monitoring Network (PMN), was first described by Feltz and others (1971) as part of the National Pesticide Monitoring Program. The PMN was implemented from late 1974 through 1981 by the U.S. Environmental Protection Agency (EPA) and the U.S. Geological Survey (USGS). The USGS collected all samples and the EPA analyzed all samples.

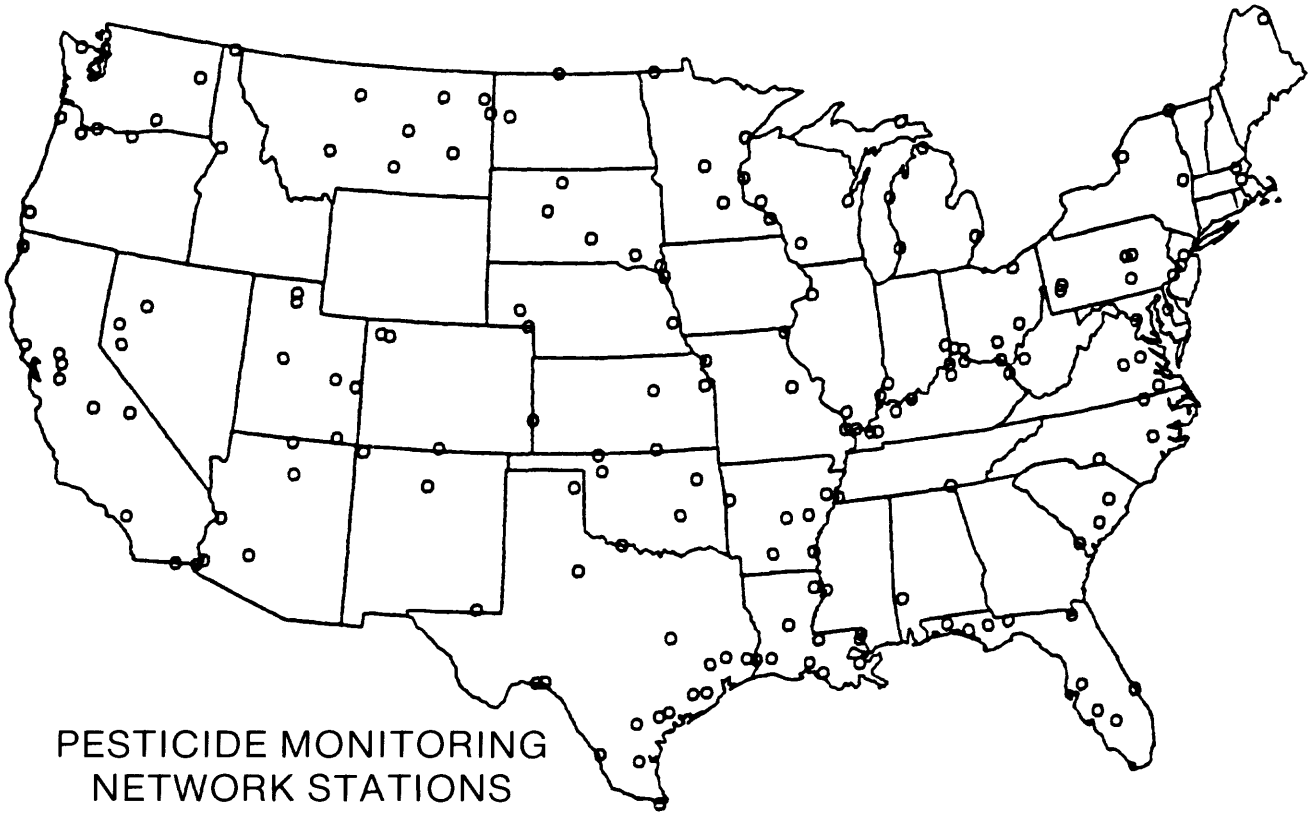
The original purposes of the program, described by Feltz and others (1971), were to provide "a continuing assessment of the general levels of pesticides in runoff and bottom sediments of the Nation's rivers" and to seek "to identify possible problem areas." The network was designed to "provide information in terms of mean levels of pesticides and their ranges of variation" and to detect "both national and regional trends" in pesticide residues in the hydrologic environment. Although the stated goals of the program have not been entirely met, the data collected allow a useful assessment of certain aspects of the distribution of pesticides in the Nation's rivers, particularly organochlorine compounds in bed sediments, and provide insights regarding the desirable features of future monitoring efforts.

The purpose of this study was to evaluate data from the PMN for the 48 contiguous States and, on the basis of this analysis and other factors, to assess implications for the scope and attributes of future monitoring efforts.

MONITORING FRAMEWORK

Sampling Strategy

The network for monitoring pesticides originally consisted of 161 stations selected from the National Stream Quality Accounting Network (NASQAN). Details of the NASQAN network are described by Ficke and Hawkinson (1975). Figure 1 shows locations of PMN stations and, for later reference, farm production regions. Station names are listed by State in appendix 1. The total number of PMN stations varied slightly over time as additions and substitutions were made. The original 161 stations of the



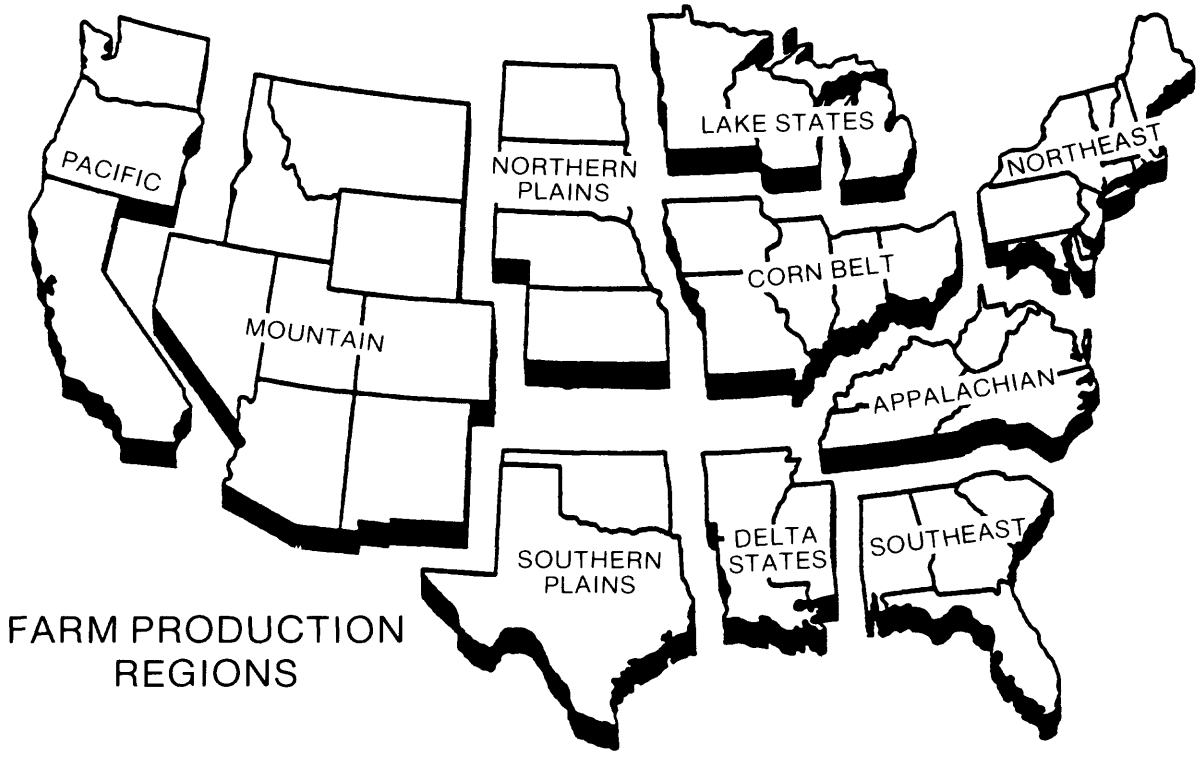


Figure 1. Locations of stations in the Pesticide Monitoring Network and farm production regions of the U.S. Department of Agriculture (farm regions map is from Andrilenas, 1974).

PMN were selected by choosing one station in each odd-numbered hydrologic accounting unit (see Ficke and Hawkinson, 1975, for description of accounting units) in the continental United States to yield an even nationwide distribution, Drainage areas of PMN stations span more than two orders of magnitude, with most in the range of 1,000 to 100,000 square miles.

Both unfiltered water samples and bed-sediment samples were collected at regular intervals at each station. Depth-integrated samples of water were collected at several verticals along the river cross section at each station and were submitted separately to the analytical laboratory, where they were then composited before analysis. Bed-sediment samples also were collected at several locations along the cross section, but in rocky reaches considerable adjustment, including moving upstream or downstream, was often necessary to obtain useful samples. Bed-sediment samples for a cross section were composited in the field. Details about sampling apparatus and technique for both water and bed sediments are described by Feltz and others (1971) and by Feltz and Culbertson (1972).

Samples were collected quarterly for water and twice a year for bed sediments. Water samples generally were collected in November, February, May, and August, and bottom-sediment samples in November and May. This schedule was a purposeful attempt to include the range of extremes of general seasonal flow conditions and expected pesticide occurrence (Feltz and others, 1971).

Chemical Analysis and Data Reporting

The presence and concentrations of pesticides in four chemical groups—organochlorine insecticides, organophosphate insecticides, triazine herbicides, and chlorophenoxy herbicides—were determined. All analyses were by gas chromatograph, with an electron capture detector used for organochlorine and chlorophenoxy compounds, a flame photometric detector for organophosphate compounds, and a nitrogen-phosphorus detector for triazine herbicides. Analytical methods, which were consistent throughout the program, were essentially the same as those described by Wershaw and others (1983).

Measurements were reported when the concentration was high enough to be considered a reliable detection of the particular compound. Prior to 1978, these data-reporting limits were determined for each analysis. In 1978, constant data-reporting limits were established by the EPA (Lucas and others, 1980). The 1978 reporting limits are also the best available estimate of pre-1978 limits, and they are the limits referred to and given in this report. Measurements falling below a reporting limit were reported as "not detected."

METHODS OF DATA ANALYSIS

A striking feature of the data collected from the PMN is that fewer than 10 percent of almost 3,000 water samples and fewer than 20 percent of almost 1,000 bed-sediment samples contained reportable concentrations of any of the pesticides. The sparsity of detected values allows very little to be inferred about the frequency distribution of concentrations of individual chemicals. The most meaningful way to analyze such data is by assessing geographic patterns and trends in the frequencies and levels of concentrations occurring above reporting limits.

The data were evaluated in three main ways:

1. Numbers of stations with detections and numbers of samples with detections were summarized and assessed for each chemical on both a national and a regional scale. For each chemical group of pesticides, the stations with the greatest number of detections were identified.
2. Correlations between frequency of detection of each pesticide and its degree of use were evaluated. Use and frequency of detection for each chemical were tabulated for each region and were assigned a rank. Kendall's test for rank correlation (Bradley, 1968) was then applied to the regional data to evaluate the significance of the relation between magnitudes of use and occurrence for each chemical.
3. General trends over time were descriptively evaluated for each chemical group by plotting frequency of detections over time on a national scale. Trends in detected concentrations were statistically evaluated for each pesticide for every station at which at least 6 bed-sediment samples or 10 water samples were analyzed and at least 2 detections were noted. Kendall's test for rank correlation (Bradley, 1968) was applied to this time series data to evaluate the presence and significance of changes in concentrations over time. Time series of measurements were plotted for selected stations and chemicals to show examples of the type of data available and the sensitivity of the trend test.

The results of these procedures were evaluated in relation to the chemical and physical properties of the compounds and to the geographic patterns and trends over time in the agricultural use of each compound. Each chemical is considered in one of three major groups—organochlorine insecticides, organophosphate insecticides, and the chlorophenoxy and triazine herbicides.

OVERVIEW OF PESTICIDE USE AND OCCURRENCE

All pesticides monitored in the PMN are synthetic chemicals that do not occur naturally and thus are found in rivers only as a result of their use, disposal, or manufacture. The greatest release of pesticides to the environment occurs in agricultural areas. In 1976, farms accounted for 65 percent of all pesticide use, including 74 percent of all herbicides (plant hormones, defoliants, and desiccants, for example) and 59 percent of all insecticides (miticides and fumigants, for example) (Eichers and others, 1978). Though information on nonfarm use is sparse, much is known about farm-use patterns.

Of all pesticides used on farms, crops accounted for about 98 percent and livestock 2 percent. In 1976, "nearly 85 percent of the crop pesticides were applied to 12 major crops: corn, cotton, wheat, sorghum, rice, other grain, soybeans, tobacco, peanuts, alfalfa, other hay and forage, and pasture and rangeland" (Eichers and others, 1978). Figure 2 shows farm use of insecticides and herbicides on these major crops from 1964 to 1982. Data for this figure are from Eichers and others (1968) for 1964, Eichers and others (1970) for 1966, Andrilenas (1974) for 1971, Eichers and others (1978) for 1976, and U.S. Department of Agriculture (1983) for 1982. The USDA's 1982 data did not include pesticide use in California, Colorado, Connecticut, Maine, Massachusetts, Nevada, New Hampshire, New Jersey, New Mexico, Oregon, Rhode Island, Utah, Vermont, West Virginia, and Wyoming. Use in these 15 states was estimated from their approximate proportional contribution in 1976 (about 8 percent of total national use), and those estimates were included in the use values for 1982 in figure 2.

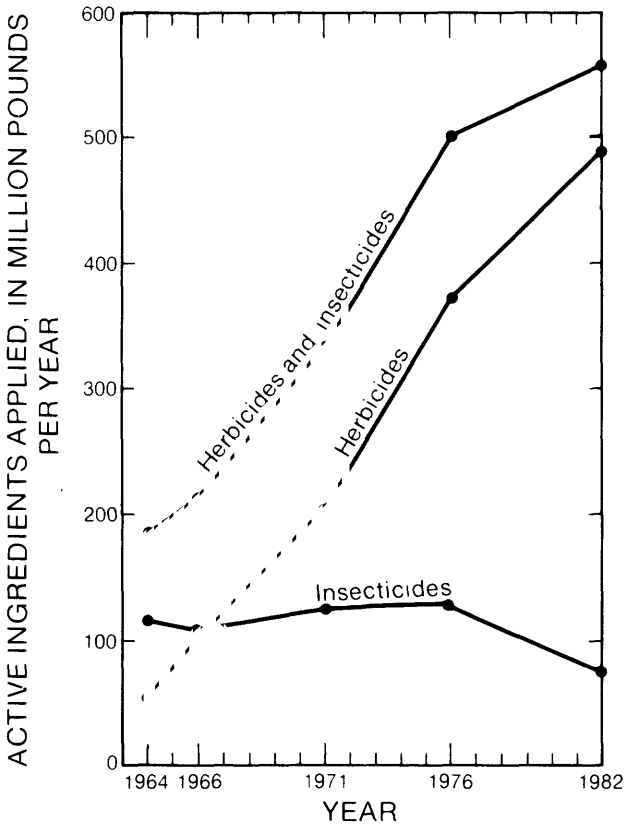


Figure 2. Trends in national use of herbicides and insecticides on major crops, 1964–82.

The overall trend, in terms of pounds applied, is one of gradually declining insecticide use and increasing herbicide use. However, much of the decline in pounds of insecticides applied, and the decline in the rate of increase in pounds of herbicides applied, are due to changes in the potency of materials used. For example, a major change in insecticide use on cotton crops has occurred since 1976. Fenvalerate and permethrin, two new chemicals requiring very low application rates, largely replaced toxaphene and methyl parathion, which were routinely applied at much higher rates (McDowell and others, 1982).

Pesticides monitored in the PMN accounted for about one-third of the total amount (by weight) of pesticides applied to major crops during the period the program was in operation. In 1976, the chlorophenoxy and triazine herbicides that were monitored accounted for 39 percent of the total amount of herbicides applied and organophosphate and organochlorine insecticides monitored accounted for 60 percent of the total amount of insecticides applied. The proportional contribution to total use of the pesticides monitored declined over the life of the program. By 1982, the herbicides monitored accounted for about 23 percent of herbicide use and the insecticides monitored accounted for about 21 percent of insecticide use.

A national overview of the numbers of detections of all pesticides monitored and the number of stations and samples is shown in tables 1 and 2 for water and bed sediments, respectively. Data reporting limits for determining detections are given later, in tables 3, 8, and 11. Tables 1 and 2 show a pattern of relatively few detections of any compounds except the triazine herbicides, diazinon, and lindane in water, relatively frequent detection of some organochlorine compounds in bed sediments, and virtually no detections of organophosphate insecticides or the herbicides in bed sediments. These general findings and more detailed analyses are further explained in the following sections. Preceeding the discussion of results for each chemical group is a summary of the

Table 1. National summary of detections of pesticides in water at Pesticide Monitoring Network stations, 1975–80

Chemical	Number of stations	Percent of stations with detections	Total number of samples	Percent of samples with detections
Organochlorine Insecticides				
Aldrin	177	2.3	2,946	0.2
Dieldrin	177	2.3	2,945	.2
Chlordane	177	.6	2,943	.0
DDD	177	4.0	2,720	.3
DDE	177	.6	2,715	.0
DDT	177	2.8	2,721	.4
Endrin	180	1.1	2,950	.1
Heptachlor epoxide.	177	4.5	2,946	.3
Lindane	177	8.5	2,945	1.1
Methoxychlor	172	.0	2,761	.0
Toxaphene	177	2.8	2,946	.4
Organophosphate Insecticides				
Diazinon	174	9.8	2,859	1.2
Ethion	174	.6	2,823	.1
Malathion	174	.6	2,859	.1
Methyl parathion.	174	2.7	2,861	.1
Methyl trithion.	174	.0	2,822	.0
Parathion	174	.6	2,856	.0
Trithion	174	1.1	2,819	.1
Triazine and Chlorophenoxy Herbicides				
Atrazine	144	24	1,363	4.8
2,4-D	186	2.4	1,764	.2
2,4,5-T	186	.6	1,765	.1
Silvex	167	.6	1,768	.1

chemical and physical properties, use, and data reporting limits for compounds in that group.

ORGANOCHLORINE INSECTICIDES

The use and selected characteristics of organochlorine insecticides are summarized in table 3. The organochlorine insecticides are generally characterized by great persistence in the natural environment, low solubility in water, and a strong tendency to adsorb to particulate matter in soil, water, and bed sediments. The most persistent are dieldrin, chlordane, and DDT, DDD, and DDE. Though table 3 shows most of the organochlorine insecticides to be nearly insoluble, lindane and toxaphene are more soluble than the others.

When some organochlorine insecticides degrade, the result is new compounds that may have either similar or different characteristics. DDT degrades to both DDE and DDD in the natural environment. As noted in table 3, this process is the only source of DDE. Aldrin degrades to dieldrin, which is more persistent than aldrin.

For most of the organochlorine insecticides, a combination of decreasing effectiveness and increasing regulatory restriction has led to a dramatic decrease in their use since the mid-1960's, as shown in

Table 2. National summary of detections of pesticides in bed sediments at Pesticide Monitoring Network stations, 1975–79

Chemical	Number of stations	Percent of stations with detections	Total number of samples	Percent of samples with detections
		Organochlorine Insecticides		
Aldrin	171	2.9	1,015	0.6
Dieldrin	172	29	1,017	12
Chlordane	171	30	1,014	9.9
DDD	171	31	990	12
DDE	172	42	989	17
DDT	171	26	992	8.5
Endrin	171	2.3	1,015	.6
Heptachlor epoxide.	171	5.3	1,017	1.0
Lindane	171	.6	1,018	.1
Methoxychlor	160	.6	941	.1
Toxaphene	171	3.5	1,014	.6
		Organophosphate Insecticides		
Diazinon	164	1.2	929	0.2
Ethion	163	.6	928	.4
Malathion	163	.0	929	.0
Methyl parathion.	163	.0	929	.0
Methyl trithion.	163	.0	928	.0
Parathion	163	.0	928	.0
Trithion	163	.0	925	.0
		Triazine and Chlorophenoxy Herbicides		
Atrazine	126	0.0	347	0.0
2,4-D	142	1.4	487	.4
2,4,5,-T	142	.7	486	.2
Silvex	142	1.4	488	.4

table 3. Overall use of organochlorine insecticides on major crops has declined from a 63-percent share of all insecticide use in 1964 to a 40-percent share in 1971 and a 28-percent share in 1976 (Eichers and others, 1970; 1978). Data for 1982 show that this share has decreased further, to less than 10 percent (U.S. Department of Agriculture, 1983). Only toxaphene retained a major share of total use through the 1970's, though by 1980 the use of toxaphene on cotton, its major application, was reduced to almost zero (McDowell and others, 1982). Despite these trends in farm use, chlordane, heptachlor, methoxychlor, and toxaphene are still used heavily off farms, as indicated in table 3 by the disparity between total use and farm use. Chlordane and heptachlor are used extensively for termite control.

Data reporting limits varied among organochlorine insecticides, making it difficult to determine whether differences among chemicals in the frequency of detection are due to their properties and use or to our ability to measure them. Reporting limits were lowest for aldrin, heptachlor epoxide, and lindane, at 0.01 µg/L, and highest for toxaphene, at 0.25 µg/L. Reporting limits for most compounds are greater than established quality criteria for water (table 3), making it difficult to evaluate the environmental significance of monitoring results. No quality criteria have been established for bed sediments.

Two different national or seminational efforts to monitor organochlorine insecticides in the Nation's river waters were made prior to the start of the PMN in 1975. Neither of the previous programs

Table 3. Selected characteristics and use of organochlorine compounds monitored in the Pesticide Monitoring Network

[µg/L, microgram per liter; nd, no available data; nr, none reported]

Chemical	Data reporting limit[1/] (ug/L)	Quality criteria[2/] (ug/L) human health	Quality criteria[2/] (ug/L) aquatic life	Solubility[3/] (µg/L)	Relative persistence[4/]	Main uses and sources	Use on farms[5/] (million pounds per year) 1966	1971	1976	1982	1981 total use[6/] (million pounds per year)
Aldrin	.01	.0007	.002	13.	Low	Corn	15 (Most farm uses canceled 1974)	7.9	0.9	nr	0.8
Dieldrin	.03	.0007	.002	22.	Medium	Termite control, degradation product of aldrin	0.7 (Most farm uses canceled 1974)	0.3	nr	nr	0
Chlordane	.15	.005	.004	56.	High	Corn, termites, general purpose	.5 (Most farm uses canceled 1974)	1.9	nr	nr	9.6
DDD	.05	.0002	.001	5.	High	Fruits and vegetables, degradation product of DDT	2.9 (Canceled 1972)	.2	nr	nr	0
DDE	.03	.0002	.001	10.	High	Degradation product of DDT and DDD	nr	nr	nr	nr	0
DDT	.05	.0002	.001	17.	High	Cotton, fruits, vegetables, general purpose	27 (Canceled 1972)	.14	nr	nr	0
Endrin	.05	.1*	.002	14.	nd	Cotton, wheat	.6	1.4	.8	nr	.3
Heptachlor epoxide.	.01	.003	.004	30.	Low	Degradation product of heptachlor which is used mainly on corn	1.5	1.2	.6	nr	2.0
Lindane	.01	.4*	.08	150.	Medium	Livestock, seed treatment, general purpose	.7	.7	.2	nr	.8
Methoxychlor	.10	100.*	.03*	3.	nd	Livestock, alfalfa, general purpose	2.6	3.0	3.8	0.6	5.0
Toxaphene	.25	.007	.013	400.	nd	Cotton, livestock	35	37	33	5.9	16

1/ Bed-sediment reporting limits are 10 times greater and in units of micrograms per kilogram (Lucas and others, 1980).

2/ Except for values marked by asterisk, which are from U.S. Environmental Protection Agency (1977), all criteria are from U.S. Environmental Protection Agency (1980). The human health criteria from the 1980 publication represent the average concentrations associated with an incremental increase in cancer risk of 10^{-5} (one additional cancer per 100,000 people) over a lifetime of exposure. The aquatic life criteria are for freshwater and are 24-hour average concentrations.

3/ Kenaga and Goring (1980).

4/ Relative persistence with organochlorine group as estimated from Wauchope (1978) and Hiltbold (1974).

5/ Data for 1966, Eichers and others (1970); for 1971, Andrilenas (1974); for 1976, Eichers and others (1978); for 1982, U.S. Department of Agriculture (1983). Data for 1982 do not include use on livestock.

6/ Mark H. Glaze, U.S. Environmental Protection Agency, written communication, 1983.

monitored bed sediments. In 1965, the U.S. Geological Survey began the Western Streams Program to monitor pesticides in rivers of the Western United States. The original network consisted of 11 stations, sampled monthly; in 1967, the number of stations was increased to 20. The network was continued until 1971. Details concerning the operation and findings of this program are reported in Manigold and Schulze (1969) and in Schulze and others (1973). In brief, one or more organochlorine insecticides (aldrin, DDD, DDE, DDT, dieldrin, endrin, heptachlor, lindane, chlordane, and toxaphene) were detected at least once at all but one station over the period of record, and the overall number of detections declined over the period of monitoring. Data reporting limits were lower than those for the PMN. The frequency of occurrence of organochlorine insecticides at concentrations greater than PMN data reporting limits decreased from an average number of detections per 100 samples of about 15 in 1968–69 to 13 in 1969–70, and to 8 by the 1970–71 water year. These frequencies were computed from the total number of detections for all 10 organochlorines divided by the number of samples collected.

The second effort to monitor the distribution of organochlorine insecticides was conducted by the Federal Water Quality Administration (FWQA) from 1964 through 1968. The effort consisted of five annual one-sample surveys for the same organochlorine chemcials monitored in the PMN, at 100 stations on major rivers of the United States. The details and results of the program are described by Lichtenberg and others (1970). Results of the FWQA program generally agreed with the U.S. Geological Survey findings for western streams. Detectable amounts of organochlorine insecticides were found to be widespread, but after peaking in 1966, detections dropped sharply in 1967 and 1968 as use declined. Data reporting limits in the FWQA program were less than those for the PMN. In the 1967–68 water year, the first year of the U.S. Geological Survey's Western Streams Program, two FWQA surveys were conducted, one in September 1967 and one in June 1968. Results of those surveys showed an average frequency of detection of organochlorine insecticides above PMN reporting limits of 14 detections per 100 samples.

Patterns of Occurrence

Results from the PMN study for organochlorine insecticides in water and bed sediments nationwide are summarized in tables 1 and 2. These results reflect the combined effects of variable data reporting limits, variable use, variable persistence and solubility, and degradation pathways which were discussed previously. The striking feature overall is the very low frequency of detection of organochlorine compounds in water samples and the moderate frequency of detection in bed sediments. The very low number of detections in water samples is consistent with the hydrophobic nature of these compounds and the decreasing trends evident in the previous monitoring studies. There are a number of more specific aspects of these data that merit discussion, however.

An obvious factor potentially affecting the frequency of occurrence of a pesticide is its rate of use. On that basis, one would expect toxaphene, methoxychlor, DDT, and aldrin to occur most frequently. This was not the case, however. Toxaphene and methoxychlor have particularly high data reporting limits (table 3) and, thus, were seldom detected. DDT degrades over time. Though it was detected fairly often in bed sediments, its degradation products, DDD (low use) and DDE (not used), were detected even more often. Aldrin, which has a low reporting limit but degrades fairly rapidly to dieldrin and endrin, was seldom detected in either water or bed sediments. Its more persistent degradation product, dieldrin, was detected in about 12 percent of bed-sediment samples despite little direct use. Thus, the patterns of detection that would be expected from pesticide use alone may not be present because of varying chemical properties and analytical capabilities.

In contrast with these heavily used compounds, lindane was used much less and yet was detected most frequently in water. The combination of lindane's relatively high solubility and persistence and a low reporting limit probably explains this. Chlordane, used only slightly more than lindane, was almost never detected in water samples but was one of the most frequently detected in bed-sediment samples. Chlordane is one of the most persistent of the organochlorine insecticides and is less than half as soluble as lindane.

The geographic distribution of detections and use of organochlorine insecticides is shown in table 4, according to the farm production regions identified in figure 1. The data for water samples show no strong relationships between detection and use, largely because of the small number of detections. Stations with the greatest number of detections in water are listed in table 5. The regional breakdown of bed-sediment data in table 4 shows moderately strong positive relationships between detections and use for chlordane, DDT, endrine, heptachlor epoxide, and toxaphene. Most of the other insecticides measured in bed sediments exhibit little or no relationship to use, in part because relatively few values are detected

Table 4. Regional patterns of detection of organochlorine insecticides in water and bed-sediment samples and their use on farms

[The upper number is the percentage frequency of detections for all samples analyzed for that region, and the lower number is the percentage of national use on farms that occurred in that region. Blanks signify no detections or use. All correlations for which the probability, p, is given are positive. The value of p is the approximate probability of incorrectly rejecting the null hypothesis that there is no correlation between use and occurrence. Values of p are not shown if $p>0.5$]

Chemical	Northeast (16 stations)	Lake States (14 stations)	Corn Belt (17 stations)	Northern Plains (12 stations)	Appalachian (18 stations)	Southeast (13 stations)	Delta States (16 stations)	Southern Plains (22 stations)	Mountain (31 stations)	Pacific (18 stations)	p for rank correlations between detection and use
Water											
Aldrin (1971 use)	—	0 1	0 94	0.4 3	0 1	0 1	0.5 <1	0.3 <1	0 <1	0.7 <1	—
Dieldrin (1971 aldrin use)	0.4 0	0 1	0.4 94	0.4 3	0 1	0 1	0 <1	0 <1	0 <1	1.3 <1	—
Chlordane (1971 use)	0 32	0 15	0 38	—	0 2	0 4	—	0 3	0 1	0.3 5	—
DDD (1971 DDT use)	0.4 2	0.9 <1	0 <1	—	0.4 7	0 50	0 34	0 6	0.2 <1	1.1 <1	—
DDE (1971 DDT use)	0 2	0 <1	0 <1	—	0 7	0 50	0 34	0 6	0.4 <1	0 <1	—
DDT (1971 use)	0 2	0 <1	0 <1	—	0 7	0 50	1.0 34	0.3 6	0.2 <1	2.1 <1	—
Endrin (1971 use)	0 <1	—	0 3	0 <1	—	0 24	0.5 70	—	0 <1	0.3 3	0.12
Heptachlor epoxide (1976 use)	—	0 2	0.8 96	—	—	0.4 0	1.0 0	0.3 2	0.2 <1	0.7 0	—
Lindane (1971 use)	0.4 3	0 1	0.4 1	1.2 1	0 <1	0.4 23	0 61	2.0 0	0.4 <1	5.3 10	—
Methoxychlor (1976 use)	0 33	—	0 11	—	0 2	0 2	—	0 2	0 <1	0 51	—
Toxaphene (1976 use)	0 1	0 <1	0 2	0.4 <1	0 8	0 49	1.9 33	0.5 1	0.8 5	—	—
Bed Sediments											
Aldrin (1971 use)	—	0 1	4.0 94	0 3	0 1	0 1	2.0 <1	0.6 <1	0 <1	0 <1	.44
Dieldrin (1971 aldrin use)	31 0	2.4 1	33 94	0 3	10 1	9.6 1	14.6 <1	5.8 <1	3.6 <1	21 <1	.47
Chlordane (1971 use)	27 32	2.3 15	14 38	1.1 0	9.1 2	11 4	8.2 0	9.7 3	2.4 1	21 5	.03
DDD (1971 ODT use)	35 2	8.4 <1	6.9 <1	—	14 7	19 50	28 34	6.1 6	4.9 <1	19 <1	.07
DDE (1971) DDT use)	33 2	9.5 <1	4.9 <1	—	18 7	24 50	28 34	21 6	12 <1	29 <1	.05
DDT (1971) use	28 2	2.4 <1	2.0 <1	—	12 7	9.8 50	20 34	4.7 6	3.0 <1	16 <1	.03
Endrin (1971 use)	0 <1	—	2.0 3	0 <1	—	1.0 24	4.1 70	—	0.6 <1	0 3	.03
Heptachlor epoxide (1976 use)	1.1 0	1.2 2	5.9 96	—	—	—	—	0.6 2	0 <1	1.4 0	.12
Lindane (1971 use)	1.1 3	0 1	0 1	0 1	0 <1	0 23	0 61	—	0 <1	0 10	—
Methoxychlor (1976 use)	0 33	—	0 11	—	0 2	0 2	—	0 2	0 <1	1.4 51	.30
Toxaphene (1976 use)	0 1	0 <1	0 2	0 <1	1.0 8	1.0 49	4.2 33	0.6 1	0.6 5	—	.03

for certain regions. However, several of these insecticides are frequently detected in regions reporting low farm use, which may indicate the importance of nonagricultural sources of these chemicals. Among the most persistent of the organochlorines, DDD, DDE, DDT, and dieldrin were detected relatively often in both the Northeast and the Pacific regions despite low 1971 use on farms in those regions (and even

Table 5. Four stations with the greatest number of detections of organochlorine insecticides in water, 1975–80 (24 samples per station)

Station	Total detections	Chemicals detected
New River at international boundary, Calexico, Calif., Pacific region.	22	9
Tensas River at Tendal, La., Delta region.	10	5
Santa Ana River below Prado Dam, Calif., Pacific region.	10	2
Red River near Burkburnett, Tex., Southern Plains region.	5	5

much lower farm use more recently). Chlordane was detected often in the Pacific States despite low 1971 use on farms and a high data reporting limit.

It may be that in the Northeast a combination of heavy population and industrialization led to extensive nonfarm use of DDD, DDT, dieldrin and (or) aldrin, and incidental release of these compounds from chemical production facilities. The 10 stations with the greatest number of detections in bed sediments are listed in table 6. Three of the Northeast region stations in table 6, the Schuylkill, Raritan, and Delaware, are downstream of or within industrialized areas. This may account for the high frequency of occurrence of these chemicals despite low agricultural use. All but 3 of the 10 stations in table 6 are located in the three regions along the East Coast.

In the Pacific region, DDT was much more heavily used in 1966 than in 1971 (Eichers and others, 1970), suggesting that heavy use prior to the 1970's may partly explain the high frequency of detection of DDT, DDD, and DDE there. Most detections of the DDT family of compounds in the Pacific region occurred at stations located within two agricultural basins, the Yakima River, Wash., and the John Day River, Oreg., and within two basins characterized by a mixture of residential and agricultural development, the Santa Ana River, Calif., and the Tualatin River, Oreg. These latter 2 stations ranked in the top 10 nationwide in number of detections of organochlorines (table 6). The Yakima, Santa Ana, and Tualatin accounted for most detections of chlordane and dieldrin in the Pacific region. Both chlordane and dieldrin were used on fruit and nut orchards and on vegetables in 1971 (Andrilenas, 1974). The Yakima, Santa Ana, and Tualatin basins contain substantial orchard areas and vegetable farms. As discussed earlier, the Santa Ana and Tualatin also contain residential areas where use of chlordane for termite control may have been significant. Thus, while there was generally light use of DDT, DDD, DDE, chlordane, and dieldrin in the Pacific region during the early 1970's, these chemicals were detected in the bed sediments of rivers draining areas where local use of these chemicals may have been heavy.

Trends in Concentration

On a national scale, concentrations of organochlorine insecticides in both water and bed sediments appear to have erratically but gradually decreased since 1975. Frequencies of detection for all stations and samples are shown for the periods of record for water and bed sediments in figure 3. Average numbers of detections per sample were computed by summing the number of detections above PMN reporting limits for all organochlorine compounds for a given year and dividing by the number of samples analyzed for organochlorines that year. Thus, the maximum number of detections per sample is 11, since 11 organochlorine insecticides were monitored.

Comparison of PMN data for water samples with data from the U.S. Geological Survey Western Streams Program indicates a marked reduction in the frequency of detections (above PMN reporting limits) since the late 1960's. For 16 stations that were identically or similarly located in the programs, the Western Streams Program showed an average frequency of detection during 1968–71 of about 12

Table 6. Ten stations with the greatest number of detections of organochlorine insecticides in bed sediments, 1975–79 (10 samples per station)

Station	Total detections	Chemicals detected
Schuylkill River at Philadelphia, Pa., Northeast region.	24	5
Black River at Kingstree, S. C., Southeast region.	23	6
Tualatin River at West Linn, Oreg., Pacific region.	23	5
Big Muddy River at Murphysboro, Ill., Corn Belt region.	22	7
Santa Ana River below Prado Dam, Calif., Pacific region.	19	5
Pee Dee River near Rockingham, N. C., Appalachian region.	17	7
Raritan River near South Bound Brook, N. J., Northeast region.	17	6
Delaware River at Trenton, N. J., Northeast region.	16	5
West Branch Susquehanna River at Lewisburg, Pa., Northeast region.	16	5
Chipola River near Altha, Fla., Southeast region.	15	6

detections per 100 samples, compared with an average of less than 1 detection per 100 samples during 1975–80 in the PMN.

Trends were also evaluated by statistically assessing trends for every station-chemical combination for which at least 2 detections and at least 10 water samples or 6 bed-sediment samples were analyzed. Trend results are shown in table 7.

Little about national or regional patterns can be inferred from water-sample data. Data were sufficient to test trends for only 13 station-chemical combinations out of almost 2,000 possible. A total of five trends were detected at the $\alpha = 0.30$ significance level—with one uptrend and four downtrends. A two-sided binomial significance test of the null hypothesis that there is an equal number of significant ($\alpha = 0.30$) trends in each direction indicated a rejection of that hypothesis at $p = 0.38$. This is only weak evidence that there are more downtrends than uptrends. Only two trends were detected at a significance level of $\alpha = 0.10$, both for the New River at the U.S. and Mexico border, California. Data for the two chemicals, aldrin and dieldrin, showing trends at the New River station are shown in figure 4. The entire drainage basin for this station is in Mexico, and no information is available on pesticide use. Of the three trends detected at other stations, two were downtrends in heptachlor and toxaphene for the Tensas River at Tendal, La., and one was a downtrend in lindane for the San Antonio River at Goliad, Tex.

Bed-sediment data were sufficient to test for trends for a total of 123 station-chemical combinations out of about 2,000 possible. As shown in table 7, a total of 36 trends were detected at the $\alpha = 0.30$ significance level, with 7 uptrends and 29 downtrends. A two-sided binomial significance test of the null hypothesis that there is an equal number of significant ($\alpha = 0.30$) trends in each direction indicated rejection of that hypothesis at $p = 0.0003$. This is strong evidence that there are more downtrends than uptrends. A total of 16 of these trends were significant at $\alpha = 0.10$—2 uptrends and 14 downtrends. Overall, the trend results in table 7 indicate that, nationally and for all but one region, trends in bed-sediment levels of organochlorine insecticides are either undetectable or predominantly downward.

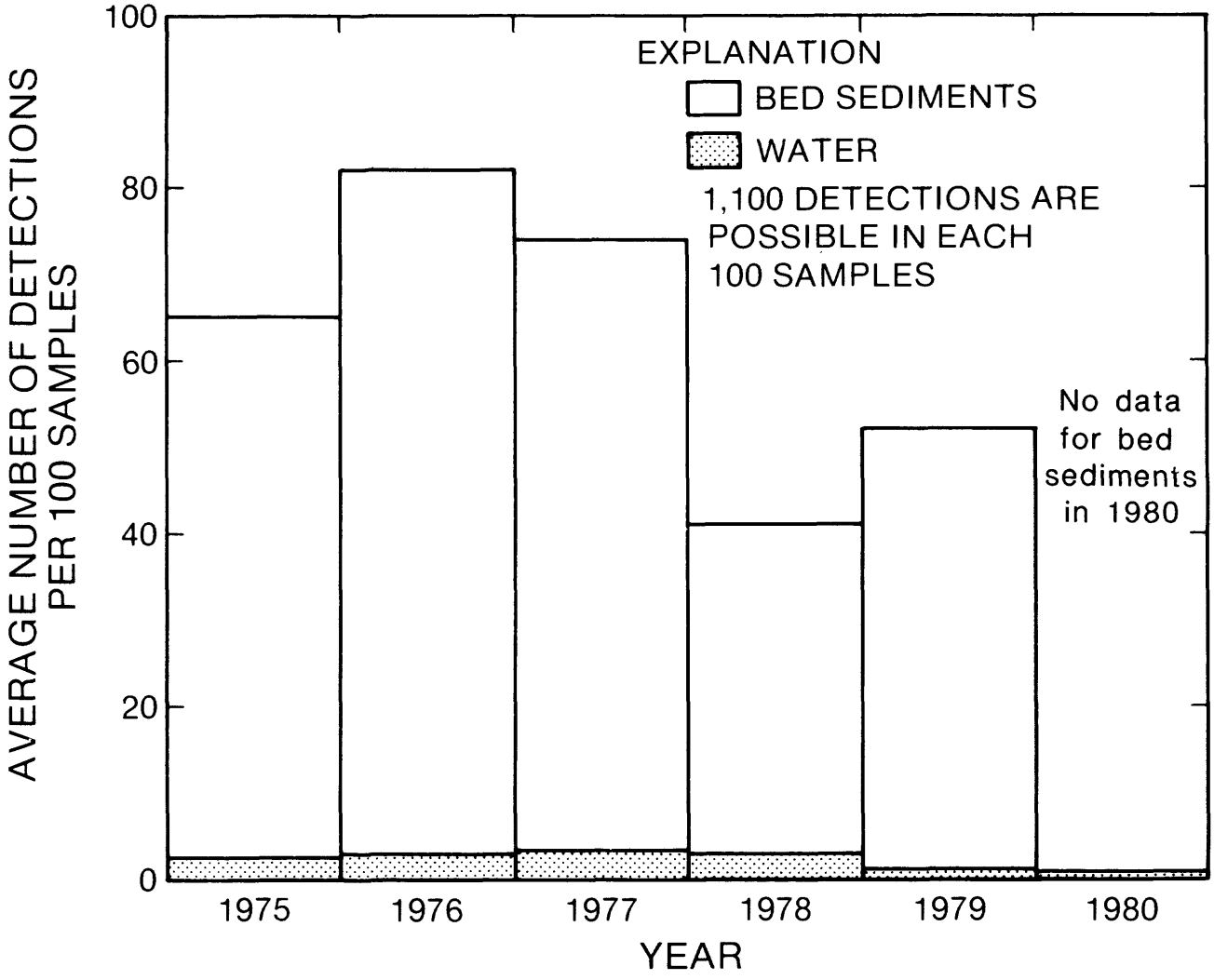


Figure 3. Frequency of detection of organochlorine insecticides in Pesticide Monitoring Network samples.

Significant trends in bed-sediment levels were concentrated at relatively few of the stations for which there were adequate data for testing trend. Trends were most often evident for the chemicals most frequently detected: DDD, DDE, DDT, chlordane, and dieldrin. Of the seven uptrends nationwide, five were at the Black River at Kingstree, S.C., which had uptrends in DDD, DDE, DDT, chlordane, and dieldrin. Of the 29 downtrends nationwide, 18 were at just 6 stations and the remaining 11 were at 11 different stations. As an example of the type of data available for bed sediments and the results of the trend test for uptrends and downtrends of different significance levels, data for dieldrin in bed sediments in the Black River, S.C., and the San Antonio River, Tex., are shown in figure 5. The San Antonio River had downtrends in the same five chemicals for which the Black River had uptrends.

ORGANOPHOSPHATE INSECTICIDES

The general characteristics and use of the organophosphate chemicals monitored in the PMN are summarized in table 8. Chemicals in the organophosphate group of insecticides, in contrast to the organochlorine chemicals, are generally short-lived (for example, most persist in soil only 1 to 12 weeks after application) and are highly soluble in water. Diazinon, the most persistent of the organophosphate compounds monitored, may last for several months.

Farm use of the organophosphate chemicals monitored has declined steadily through the 1970's, though not as dramatically as the organochlorine insecticides (table 8). Methyl parathion was used most, mainly on cotton. Some of the chemicals monitored—ethion, methyl trithion, and trithion—were

Table 7. Trends in concentration of organochlorine insecticides in water and bed sediments

[$\frac{0}{2}{}^{1}$ signifies no stations with uptrends, two stations with downtrends, and one station with no trend. Trends were tested for each station with at least 10 observations and at least 2 detections. The minimum significance level for identification of trend was $\alpha = 0.30$]

Chemical	Northeast (16 stations)	Lake States (14 stations)	Corn Belt (17 stations)	Northern Plains (12 stations)	Appalachian (18 stations)	Southeast (13 stations)	Delta States (16 stations)	Southern Plains (22 stations)	Mountain (31 stations)	Pacific (18 stations)	National (177 stations)
Water											
Aldrin	–	–	–	–	–	–	–	–	–	$\frac{1\ 0}{0}$	$\frac{1\ 0}{0}$
Dieldrin	–	–	–	–	–	–	–	–	–	$\frac{0\ 0}{1}$	$\frac{0\ 0}{1}$
Chlordane	–	–	–	–	–	–	–	–	–	–	–
DDD	–	–	–	–	–	–	–	–	–	$\frac{0\ 1}{0}$	$\frac{0\ 1}{0}$
DDE	–	–	–	–	–	–	–	–	–	–	–
DDT	–	–	–	–	–	–	$\frac{0\ 1}{0}$	–	–	$\frac{0\ 1}{0}$	$\frac{0\ 2}{0}$
Endrin	–	–	–	–	–	–	–	–	–	–	–
Heptachlor epoxide	–	–	–	–	–	–	$\frac{0\ 0}{1}$	–	–	–	$\frac{0\ 0}{1}$
Lindane	–	–	–	$\frac{0\ 1}{0}$	–	–	–	$\frac{0\ 1}{1}$	–	$\frac{0\ 2}{0}$	$\frac{0\ 4}{1}$
Methoxychlor	–	–	–	–	–	–	–	–	–	–	–
Toxaphene	–	–	–	–	–	–	$\frac{0\ 0}{1}$	–	$\frac{0\ 1}{0}$	–	$\frac{0\ 1}{1}$
All compounds	–	–	–	$\frac{0\ 1}{0}$	–	–	$\frac{0\ 1}{2}$	$\frac{0\ 1}{1}$	$\frac{0\ 1}{0}$	$\frac{1\ 4}{1}$	$\frac{1\ 8}{4}$
Bed Sediment											
Aldrin	–		$\frac{0\ 1}{0}$	–	–	–	–	–	–	–	$\frac{0\ 1}{0}$
Dieldrin	$\frac{0\ 4}{2}$	–	$\frac{0\ 3}{4}$	–	$\frac{0\ 2}{0}$	$\frac{1\ 1}{0}$	$\frac{0\ 1}{0}$	$\frac{0\ 0}{1}$	$\frac{0\ 1}{1}$	$\frac{0\ 2}{0}$	$\frac{1\ 14}{8}$
Chlordane	$\frac{0\ 1}{3}$	$\frac{0\ 1}{0}$	$\frac{0\ 2}{0}$	–	$\frac{0\ 0}{1}$	$\frac{1\ 2}{1}$	–	$\frac{0\ 1}{2}$	$\frac{0\ 1}{0}$	$\frac{0\ 2}{0}$	$\frac{1\ 10}{7}$
DDD	$\frac{0\ 7}{1}$	$\frac{0\ 2}{0}$	$\frac{0\ 2}{0}$	–	$\frac{0\ 4}{0}$	$\frac{1\ 2}{1}$	$\frac{0\ 1}{0}$	$\frac{0\ 0}{2}$	$\frac{0\ 3}{0}$	$\frac{0\ 2}{0}$	$\frac{1\ 23}{4}$
DDE	$\frac{0\ 6}{1}$	$\frac{0\ 2}{0}$	$\frac{0\ 0}{1}$	–	$\frac{0\ 3}{2}$	$\frac{1\ 3}{1}$	$\frac{0\ 1}{0}$	$\frac{0\ 4}{2}$	$\frac{0\ 3}{0}$	$\frac{0\ 3}{0}$	$\frac{1\ 25}{7}$
DDT	$\frac{0\ 6}{0}$	–	–	–	$\frac{0\ 3}{0}$	$\frac{1\ 1}{0}$	$\frac{1\ 0}{0}$	$\frac{1\ 0}{2}$	–	$\frac{0\ 2}{0}$	$\frac{3\ 12}{2}$
Endrin	–	–	$\frac{0\ 0}{1}$	–	–	$\frac{0\ 1}{0}$	–	–	–	–	$\frac{0\ 1}{1}$
Heptachlor epoxide	–	–	$\frac{0\ 1}{0}$	–	–	–	–	–	–	–	$\frac{0\ 1}{0}$
Lindane	–	–	–	–	–	–	–	–	–	–	–
Methoxychlor	–	–	–	–	–	–	–	–	–	–	–
Toxaphene	–	–	–	–	–	–	–	–	–	–	–
All compounds	$\frac{0\ 24}{7}$	$\frac{0\ 5}{0}$	$\frac{0\ 9}{6}$	–	$\frac{0\ 12}{3}$	$\frac{5\ 10}{3}$	$\frac{1\ 3}{0}$	$\frac{1\ 5}{9}$	$\frac{0\ 8}{1}$	$\frac{0\ 11}{0}$	$\frac{7\ 87}{29}$

used very little on farms during the life of the PMN, though nonfarm use of ethion has been quite high.

Data reporting limits were generally higher for organophosphate compounds than for the organochlorines. Comparison of tables 3 and 8 indicates that reporting limits for organophosphate compounds are mostly in the range of 5 to 50 times greater than those for the organochlorine compounds. Within the group, diazinon had the lowest reporting limit at 0.1 µg/L. Water-quality criteria have been established for only two of the compounds, malathion and parathion, and only for

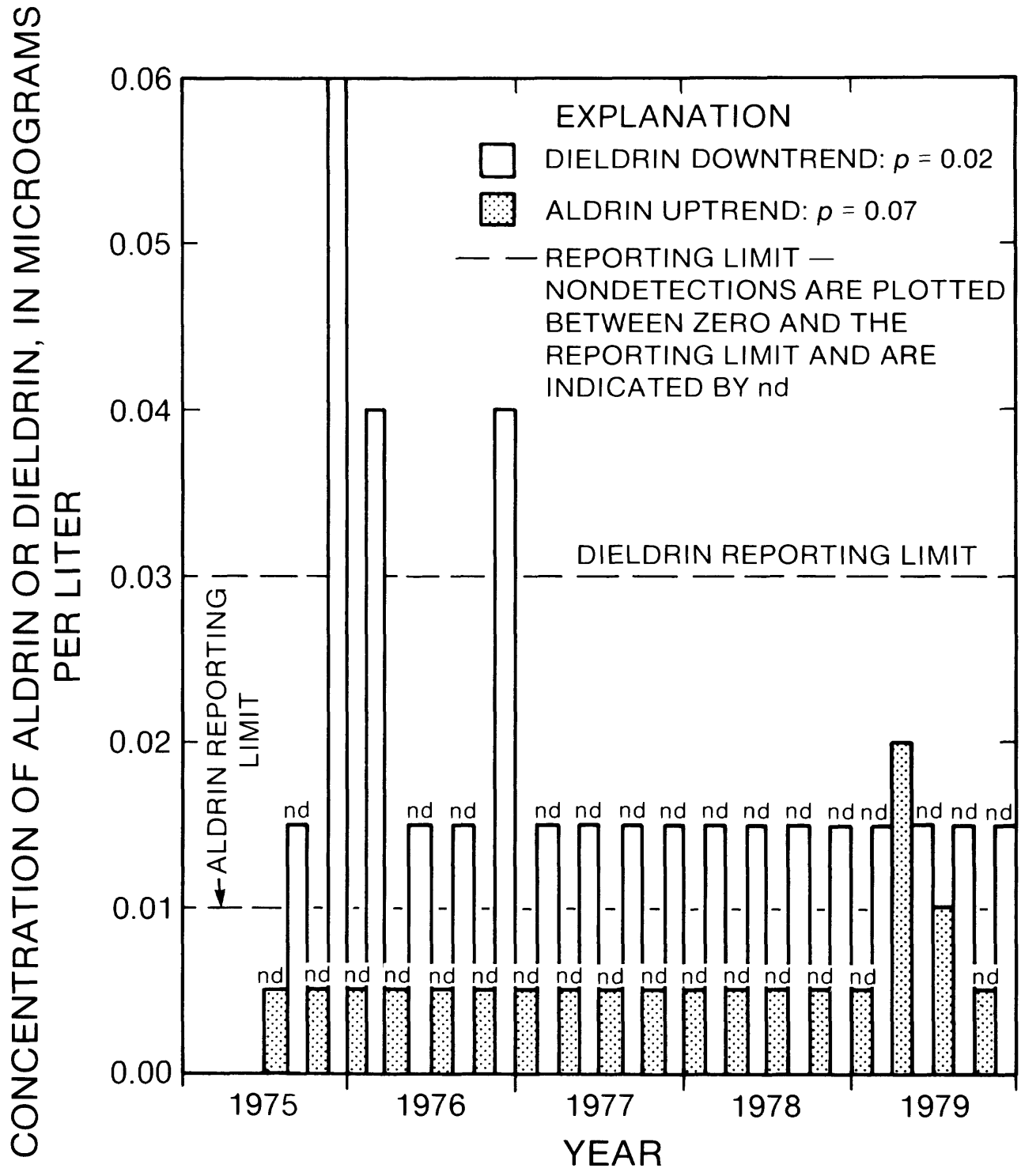


Figure 4. Aldrin and dieldrin concentrations in water samples from the New River at the border between the United States and Mexico, California.

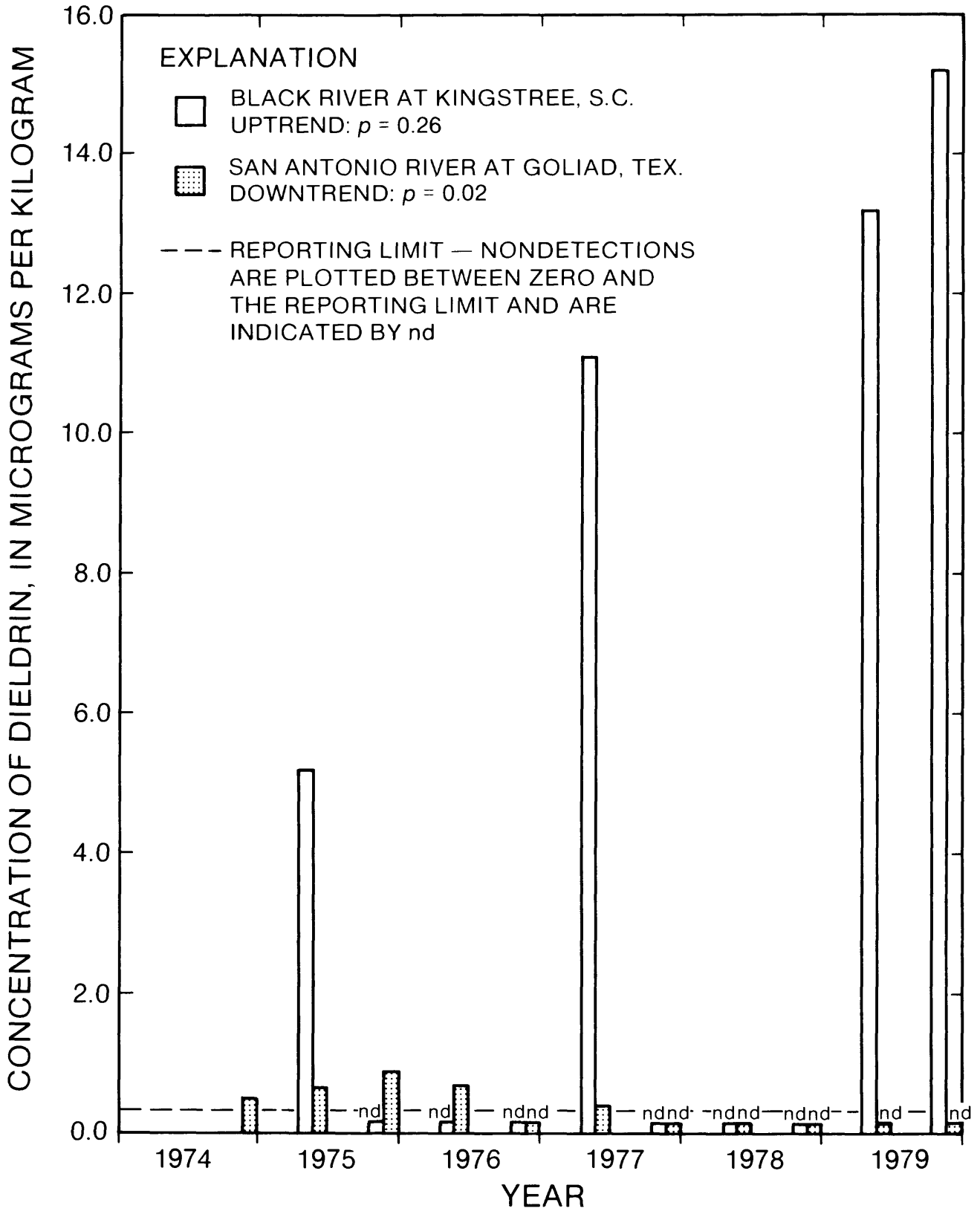


Figure 5. Dieldrin concentrations in bed sediments of the Black River, S.C., and the San Antonio River, Tex.

Table 8. Selected characteristics and use of organophosphate insecticides included in the Pesticide Monitoring Network

[µg/L, microgram per liter; nd, no available data; nr, none reported]

Chemical	Data reporting limit[1] (µg/L)	Quality criteria[2] (µg/L)		Solubility[3] (µg/L)	Relative persistence[4]	Main uses and sources	Use on farms[5] (million pounds per year)				1981 total use[6] (million pounds per year)
		human health	aquatic life				1966	1971	1976	1982	
Diazinon	.10	nd	nd	40,000	High	Corn, general purpose	5.6	3.2	1.6	0.3	9.0
Ethion	.25	nd	nd	2,000	nd	Citrus fruits	2.0	2.3	nr	nr	2.0
Malathion	.25	nd	.1	145,000	Low	General purpose	5.2	3.6	2.8	1.6	28
Methyl parathion	.25	nd	nd	57,000	Low	Cotton and wheat	8.0	28.	23.	11.0	20
Methyl trithion	.50	nd	nd	nd	nd	Not identified	nr	nr	nr	nr	0.1
Parathion	.25	nd	.04	24,000	Low	Wheat, corn, sorghum	8.5	9.5	6.6	4.0	5.0
Trithion	.50	nd	nd	340	nd	General purpose	nr	nr	nr	nr	0.1

1/ Bed-sediment reporting limits are 10 times greater and in units of micrograms per kilogram (Lucas and others, 1980).

2/ Environmental Protection Agency (1977).

3/ Kenaga and Goring (1980).

4/ Relative persistence within organophosphorus group as estimated from Wauchope (1978) and Hiltbold (1974).

5/ Data for 1966, Eichers and others (1970); for 1971, Andrilenas (1974); for 1976, Eichers and others (1978); for 1982, U.S. Department of Agriculture (1983). Data for 1982 do not include use on livestock.

6/ Mark H. Glaze, U.S. Environmental Protection Agency, written communication, 1983.

aquatic life. For both compounds, the aquatic life criterion is less than the data reporting limit.

Of the two previous monitoring studies discussed in relation to organochlorine insecticides, the USGS analyzed samples for methyl parathion, parathion, and diazinon during only the last 2 years of study (1970–71), and the FWQA analyzed for methyl parathion, parathion, ethion, malathion, and trithion in 1967 and 1968. Diazinon was detected twice and parathion once above PMN reporting limits in the USGS study; parathion and ethion were detected once in the FWQA study.

Patterns of Occurrence

National results from the PMN for organophosphate insecticides in water and bed sediments are summarized in tables 1 and 2. As with the organochlorine insecticides, the results in tables 1 and 2 reflect the combined effects of variable detection limits, degree of use, solubility, and persistence. The low frequency of detections appears to result primarily from the relatively high data reporting limits for these chemicals and their low persistence. As with toxaphene, which was heavily used but seldom detected because of the high reporting limit, methyl parathion was the most heavily used organophosphate insecticide and yet was detected only three times out of 2,861 water samples, once each at three different stations. Other chemicals in the group with equal or higher reporting limits than methyl parathion, and less use, were detected even fewer times in water. Diazinon was detected substantially more often than the other organophosphate chemicals in water, 34 detections in 2,859 samples, both because the reporting limit for diazinon is less than half that for the other chemicals and because diazinon is more persistent than the other organophosphate compounds. Owing to their high solubility in water and their low persistence, virtually all organophosphate chemicals were detected even less often in bed sediments than in water.

The geographic distribution of detections and use of organophosphate insecticides is shown in table 9, according to the farm production regions identified in figure 1. Rank correlations between detection and use are generally weak. The lack of detections, particularly for bed sediments, hampers any meaningful analysis of geographic patterns of occurrence. The six stations with more than one detection in water are listed in table 10. The one station with over a third of all detections was the New River, which drains Mexican land for which Pacific region use data do not apply.

Trends in Concentration

No trends are evident in detections of organophosphate insecticides on either a national or a regional scale or at any individual station for either water or bed sediments. Detections were too few to allow any analysis of trends in bed-sediment concentrations. There were only six bed-sediment detections at a total of three stations. A national summary of detections in water samples over time is shown in figure 6. No clear pattern is evident. Most detections were scattered within a factor of two of the detection limit. On a station-by-station basis for all organophosphate chemicals, there were sufficient numbers of detections to test for trends for only seven station-chemical combinations. No trends were evident at a significance level of $\alpha = 0.30$.

CHLOROPHENOXY AND TRIAZINE HERBICIDES

Table 11 summarizes the use and selected characteristics of herbicides monitored in the PMN. The herbicides included in the program, which include the major representatives of the chlorophenoxy and triazine groups, are generally intermediate in persistence between organochlorine and organophosphate insecticides and are highly soluble in water. Triazine herbicides are generally more persistent and less soluble than chlorophenoxy herbicides. Atrazine may persist for almost a year in soil, for example, whereas 2,4,5-T and 2,4-D usually last less than 6 months.

As figure 2 shows, the use of herbicides has rapidly increased during the past 20 years. The use of atrazine and 2,4-D accounts for much of this use. From 1971 to 1976, these two chemicals accounted for about 50 percent of all herbicide use. However, the dominance of these chemicals had begun to decrease somewhat by 1980. For example, atrazine fell from 41 percent of total herbicide application on corn in 1976 (Eichers and others, 1978) to 33 percent in 1980 (Hanthorn and others, 1982). Of the two previous pesticide monitoring efforts already discussed, only the U.S. Geological Survey study of western streams included herbicides, and only the chlorophenoxy herbicides were determined. From 1967 to 1971, no concentrations of herbicides were measured that met or exceeded the PMN data reporting limits, though there were numerous detections below the PMN limits.

Patterns of Occurrence

Results in tables 1 and 2 show virtually no detections of herbicides in bed sediments and, except

Table 9. Regional patterns of detection of organophosphate insecticides and their use on farms

[The upper number in each box is the percentage frequency of detections for all samples analyzed for that region, and the lower number is the percentage of national use on farms that occurred in that region. Blank boxes signify no detections or use. All correlations for which the probability, p, is given are positive. The value of p is the approximate probability of incorrectly rejecting the null hypothesis that there is no correlation between use and occurrence. Values of p are not shown if $p>0.5$]

Chemical	Northeast (16 stations)	Lake States (14 stations)	Corn Belt (17 stations)	Northern Plains (12 stations)	Appalachian (18 stations)	Southeast (13 stations)	Delta States (16 stations)	Southern Plains (22 stations)	Mountain (31 stations)	Pacific (18 stations)	p for rank correlations between use and detection
Water											
Diazinon (1976 use)	0 < 1	0.4 13	0.8 42	0.4 0	1.1 2	0 < 1	2.5 0	1.8 4	0.4 4	4.0 35	.32
Ethion (1971 use)	0 2	—	—	—	0 < 1	0.8 72	0 1	0 4	0 1	0 22	.16
Malathion (1976 use)	0 23	—	0 12	0 26	0 6	0 4	0 6	0 19	0 2	1.3 1	—
Methyl parathion (1976 use)	0 < 1	0 < 1	0 1	0 1	0 5	0 28	0.5 55	0.3 5	0.4 4	0 1	.32
Methyl trithion (1976 use)	—	—	—	—	—	—	—	—	—	—	—
Parathion (1976 use)	0 < 1	0 < 1	—	0 17	0 1	0 8	0 < 1	0.3 64	0 6	0 3	.16
Trithion (1976 use)	—	—	—	—	0.4 0	—	—	—	—	0.3 0	—
Bed Sediments											
Diazinon (1976 use)	0 < 1	0 13	2.0 42	—	0 2	0 < 1	—	0 4	0 4	0 35	.16
Ethion (1971 use)	0 2	—	—	—	0 < 1	4.0 72	0 1	0 4	0 1	0 22	.16
Malathion (1976 use)	0 23	—	0 12	0 26	0 6	0 4	0 6	0 19	0 2	0 1	—
Methyl parathion (1976 use)	0 < 1	0 < 1	0 1	0 1	0 5	0 28	0 55	0 5	0 4	0 1	—
Methyl trithion (1976 use)	—	—	—	—	—	—	—	—	—	—	—
Parathion (1976 use)	0 < 1	0 < 1	—	0 17	0 1	0 8	0 < 1	0 64	0 6	0 3	—
Trithion (1976 use)	—	—	—	—	—	—	—	—	—	—	—

for atrazine, few detections in water samples. For most stations, there were only 3 years of record for herbicides: 1976–78. The second most detected herbicide was 2,4-D. These results may be explained by the extremely heavy use of both atrazine and 2,4-D combined with the much greater persistence of atrazine. If data reporting limits had been lower, there probably would have been a much higher frequency of detection of both of these chemicals, and of the other herbicides as well.

Regional patterns of herbicide detections are shown in relation to regional use in table 12. The only significant rank correlation between use and detection was found for atrazine in water. Of a total of 74 herbicide detections during operation of the PMN, 48 were of atrazine. Stations at which atrazine was detected more than twice are given in table 13. All stations in table 13 are located downriver from major corn-growing areas, where virtually all atrazine is applied.

Trends in Concentration

The generally low rate of detections of herbicides and the limited time span of data available for atrazine made it impossible to evaluate trends in any herbicide concentrations in either bed sediments or water.

SUMMARY

The Pesticide Monitoring Network (PMN) for rivers was operated from 1975 to 1980. Water sam-

Table 10. Six stations with more than one detection of organophosphate insecticides in water, 1975–80 (24 samples per station)

Station	Total detections	Chemicals detected
New River at international boundary, Calexico, Calif., Pacific region.	16	3
San Antonio River at Goliad, Tex., Southern Plains region.	6	2
Gila River above diversions at Gillespie Dam, Ariz., Mountain region.	3	2
Main Canal at Vero Beach, Fla., Southeast region.	2	1
Mississippi River near St. Francisville, La., Delta region.	2	1
Santa Ana River below Prado Dam, Calif., Pacific region	2	1

ples were taken four times per year and bed-sediment samples two times per year at 160–180 stations (depending on the year) on major rivers in the United States. Samples were analyzed for 11 chlorinated hydrocarbon insecticides, 7 organophosphate insecticides, and 4 herbicides. The pesticides that were monitored accounted for about one-third of the total amount (by weight) of all pesticides applied to major crops during 1975–80.

During the life of the program, fewer than 10 percent of almost 3,000 water samples and fewer than 20 percent of almost 1,000 bed-sediment samples contained reportable concentrations of any of the compounds. The patterns of detection result from a combination of widely variable detection capabilities, chemical properties, and use. Detection limits for organochlorine compounds ranged from 0.01 to 0.25 µg/L and for organophosphate com-

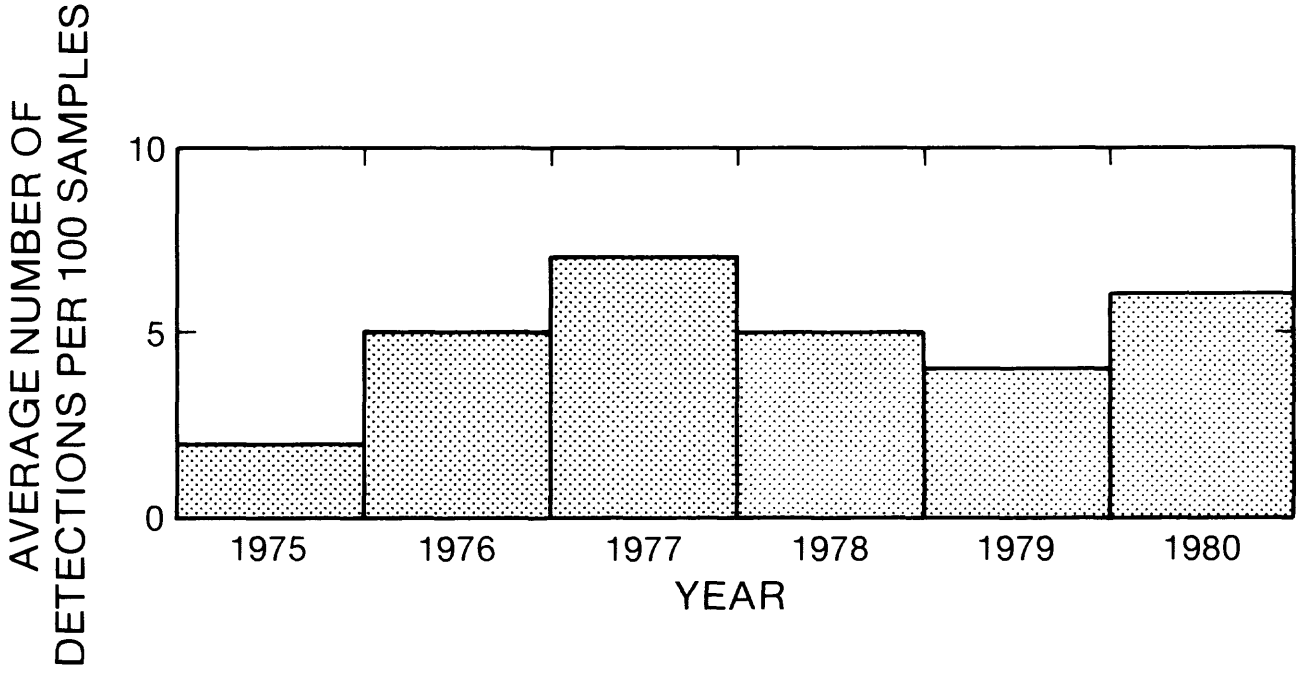


Figure 6. Frequency of detection of organophosphate insecticides in Pesticide Monitoring Network water samples.

Table 11. Selected characteristics and use of herbicides included in the Pesticide Monitoring Network

[µg/L, microgram per liter; nd, no available data; nr, none reported]

Chemical	Data Reporting limit[1] (µg/L)	Quality criteria[2] (µg/L) human health	Quality criteria[2] (µg/L) aquatic life	Solubility[3] (µg/L)	Relative Persistence[4]	Main uses and sources	Use on farms[5] (million pounds per year) 1966	1971	1976	1982	1981 total use[6] (million pounds per year)
2,4-D	.5	100	nd	900,000	Low	Wheat, rangeland, general purpose	4	31	38	23	60
2,4,5-T	.5	10	nd	240,000	Medium	Rice, rangeland, general purpose	.8	nr	nr	.2	2.2
Silvex	.5	nd	nd	140,000	nd	Sugarcane, rice, rangeland	nr	nr	nr	nr	.4
Atrazine	.5	nd	nd	33,000	High	Corn	24	54	90	76	92

1/ Bed-sediment reporting limits are 10 times greater and in units of micrograms per kilogram (Lucas and others, 1980).

2/ Environmental Protection Agency (1977).

3/ Kenega and Goring (1980).

4/ Relative persistence within herbicide group as estimated from Wauchope (1978) and Hiltbold (1974).

5/ Data for 1966, Eichers and others (1970); for 1971, Andrilenas (1974); for 1976, Eichers and others (1978); for 1982, U.S. Department of Agriculture (1983).

6/ Mark H. Glaze, U.S. Environmental Protection Agency, written communication, 1983.

Table 12. Regional patterns of detection of herbicides and their use on farms

[The upper number in each box is the percentage frequency of detections for all samples analyzed for that region, and the lower number is the percentage of national use on farms that occurred in that region. Blank boxes signify no detections or use, and na signifies no available data. All correlations for which p is given are positive. The value of p is the approximate probability of incorrectly rejecting the null hypothesis that there is no correlation between use and occurrence. Values of p are not shown if $p>0.5$]

Chemical	Northeast (16 stations)	Lake States (14 stations)	Corn Belt (17 stations)	Northern Plains (12 stations)	Appalachian (18 stations)	Southeast (13 stations)	Delta States (16 stations)	Southern Plains (22 stations)	Mountain (31 stations)	Pacific (18 stations)	p for rank correlation between use and detection
Water											
2,4-D (1976 use)	0 1	0 9	0.6 20	0.8 32	0.6 2	0 1	0.7 1	0 8	0 12	0 14	.36
2,4,5-T (1971 use)	0 1	0 2	0 6	0 17	0 1	0 1	1.4 11	0 42	0 3	0 15	—
Silvex	0 na	0 na	0 na	0 na	0 na	0 na	0 na	0 na	0 na	0.5 na	na
Atrazine (1976 use)	1.5 6	1.7 16	20 47	10 16	8.5 9	0 4	4.5 1	2.8 2	0.9 < 1	0.7 < 1	.05
Simazine (1976 use)	0 36	0 2	0 30	0 4	0 28	0 < 1	—	—	—	3.0 < 1	—
Bed sediments											
2,4-D (1976 use)	0 1	0 9	0 20	0 32	0 2	1.8 1	0 1	0 8	1.1 12	0 14	—
2,4,5-T (1971 use)	0 1	0 2	0 6	0 17	0 1	1.8 1	0 11	0 42	0 3	0 15	—
Silvex	0 na	0 na	0 na	0 na	0 na	1.8 na	0 na	0 na	1.1 na	0 na	na
Atrazine (1976 use)	0 6	0 16	0 47	0 16	0 9	0 4	0 1	0 2	0 < 1	0 < 1	—
Simazine (1976 use)	0 36	0 2	0 30	0 4	0 28	0 < 1	—	—	—	0 < 1	—

pounds from 0.10 to 0.50 µg/L; all herbicides had a detection limit of 0.50 µg/L. Most detections in water samples were of relatively persistent yet soluble compounds: atrazine (4.8 percent of samples), simazine (1.8), diazinon (1.2), and lindane (1.1). Most detections in bed-sediment samples were of the hydrophobic and persistent insecticides: DDE (17 percent of samples), DDD (12), dieldrin (12), chlordane (9.9), and DDT (8.5). Geographic patterns of detection were correlated ($\alpha = 0.10$) with use on farms for only atrazine in water, and only endrin, heptachlor, epoxide, and toxaphene in bed sediments.

Detections of organochlorine insecticides in both water and bed sediments appear to have decreased erratically but gradually during 1975–80, and levels found in selected western streams appear to be much lower than those found in a 1968–71 study. For the 1975–79 period, more stations had downtrends in bed-sediment levels than had uptrends. No clear trends were evident in concentrations of organophosphate insecticides in either water or bed sediments, and herbicide data were for too short a period of time to allow trend evaluation.

IMPLICATIONS FOR FUTURE MONITORING

The monitoring and assessment of levels and trends of pesticides in the Nation's streams and rivers is hampered by several problems, some of which are unique in comparison to other water-quality constituents. The following key problems were identified from the results of this study:

- The amounts and types of pesticides used are constantly changing over time.

Future programs aimed at monitoring and assessing pesticides in the Nation's water need to be

Table 13. Six stations with more than two detections of atrazine in water, 1976–78 (12 samples per station)

Station	Detections	Station	Detections
Kansas River at DeSoto, Kan., Northern Plains region.	8	Big Muddy River at Murphysboro Ill., Corn Belt region.	4
Mississippi River at Memphis, Tenn., Appalachian region	5	Mississippi River at Thebes, Ill., Corn Belt region.	4
Missouri River at Hermann, Mo., Corn Belt region.	5	Des Moines River at St. Francisville, Mo., Corn Belt region.	3

modified periodically to account for changing patterns in pesticide use. New chemicals need to be added as they come into use and short-lived chemicals that are no longer used can be discontinued. Some persistent chemicals, such as the organochlorine insecticides, may need to be monitored for several years after use has been discontinued.

- The use of most pesticides conforms to strong geographic patterns—what is important in one place is not in another.

Monitoring programs can evolve toward more intense sampling and analysis for the most important compounds in a region and less intense effort for infrequently used chemicals that are seldom present. This principle must be applied with caution, however. Decisions concerning which compounds to monitor should not be made from use data alone because use data may not reflect all important sources of some chemicals. Use data should be supplemented and confirmed by reconnaissance monitoring data.

- Different pesticides have markedly different chemical properties that result in different environmental behavior. For example, some are soluble and some are not, some are short-lived and some are very persistent. Little is known about the effect of these variable properties on the occurrence and transport of different pesticides in river systems. In addition, applications of pesticides tend to be very seasonal, with the application time varying by chemical, by crop, by geographic region, and by weather.

Different sampling approaches are needed for different chemicals. For example, persistent hydrophobic compounds, such as DDT or dioxin isomers, may be effectively monitored by periodic bed-sediment analysis on a fairly crude spatial scale, while soluble, short-lived compounds may require frequent water-column sampling near application sites only during application seasons. Although sampling approaches could ideally be tailored to specific groups of chemicals in a region, our lack of knowledge about occurrence and transport in relation to chemical properties makes it necessary to carry out some rather detailed studies before enough is known to understand the efficiency and meaning of a particular monitoring approach.

- Little is known about the long-term adverse effects of low levels of pesticides on aquatic ecosystems or humans.

The lack of knowledge about long-term effects of low concentrations on aquatic ecosystems and humans means that there is a strong need for studying the occurrence and distribution of pesticides at the lowest levels possible so that the presence or absence of such effects eventually may be assessed empirically.

- Pesticides are difficult to sample, identify, and measure in dilute surface water, and chemical analyses are very expensive. Some pesticide compounds are unstable and may change rapidly in sample containers prior to analysis. Detection capability varies among compounds, and identification and measurement reliability are poorly characterized.

The combination of expense and uncertainty associated with pesticide sampling and chemical analysis presents a fundamental and very difficult problem for effective monitoring and research. There is a great need for careful quality assurance involving replicate samples and "overmeasuring" by intensive sampling in both time and space in order to characterize the variablility of measured pesticide concentrations. Though the expense of up to several hundred dollars per replicate sample may strain the most sturdy of budgets, if quality assurance efforts are eliminated, the quality of data obtained will be unknown. This also implies that pesticide monitoring efforts may initally have to concentrate much effort at a few sites in order to characterize and refine sampling and analytical capabilities under field con-

ditions, and then gradually expand to greater spatial coverage as measurement capabilities are better understood.

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APPENDIX 1: PESTICIDE MONITORING STATIONS, BY STATE

Station No.	Station Name	State
02469762	Tombigbee River below Coffeeville Lock and Dam, Ala.	ALABAMA
09380000	Colorado River at Lees Ferry, Ariz.	ARIZONA
09401200	Little Colorado River at Cameron, Ariz.	ARIZONA
09426600	Bill Williams River near Planet, Ariz.	ARIZONA
09518000	Gila River above Diversions at Gillespie Dam, Ariz.	ARIZONA
09520700	Gila River near mouth, near Yuma, Ariz.	ARIZONA
09522000	Colorado River at Nib Ab Morelos Dam near Andrade, Calif.	ARIZONA
07047800	St. Francis River at Parkin, Ark.	ARKANSAS
07077800	White River at Clarendon, Ark.	ARKANSAS
07250550	Arkansas River at Dam No. 13, near Van Buren, Ark.	ARKANSAS
07263620	Arkansas River at David D. Terry Lock and Dam below Little Rock, Ark.	ARKANSAS
07265450	Missippippi River near Arkansas City, Ark.	ARKANSAS
07362000	Ouachita River at Camden, Ark.	ARKANSAS
09429490	Colorado River above Imperial Dam, Ariz.-Calif.	CALIFORNIA
10254970	New River at International Body, Calexico, Calif.	CALIFORNIA
10277400	Owens River below Tinemaha Reservoir near Big Pine, Calif.	CALIFORNIA
11074000	Santa Ana River below Prado Dam, Calif.	CALIFORNIA
11250000	Friant-Kern Canal at Friant, Calif.	CALIFORNIA
11303500	San Joaquin River near Vernalis, Calif.	CALIFORNIA
11325500	Mokelumne River at Woodbridge, Calif.	CALIFORNIA
11447650	Sacramento River at Freeport, Calif.	CALIFORNIA
11467000	Russian River near Guerneville, Calif.	CALIFORNIA
11530500	Klamath River near Klamath, Calif.	CALIFORNIA
06764000	South Platte River at Julesburg, Col.	COLORADO
08251500	Rio Grande River near Lobatos, Col.	COLORADO
09251000	Yampa River near Maybell, Col.	COLORADO
09260000	Little Snake River near Lily, Col.	COLORADO
01184000	Connecticut River at Thompsonville, Conn.	CONNECTICUT
01205500	Housatonic River at Stevenson, Conn.	CONNECTICUT
01646580	Potomac River at Chain Bridge, at Washington, D.C.	DIST. OF COL.
02231000	St. Mary's River near MacClenny, Fla.	FLORIDA
02253000	Main Canal at Vero Beach, Fla.	FLORIDA
02273000	Kissimmee River at S-65E near Okeechobee, Fla.	FLORIDA
02292480	Caloosahatchee Canal at Ortona Lock near La Belle, Fla.	FLORIDA
02296750	Peace River at Arcadia, Fla.	FLORIDA
02303000	Hillsborough River near Zephyr Hills, Fla.	FLORIDA
02329000	Ochlockonee River near Havana, Fla.	FLORIDA
02359000	Chipola River near Altha, Fla.	FLORIDA
02366500	Choctawhatchee River near Bruce, Fla.	FLORIDA
02368000	Yellow River at Milligan, Fla.	FLORIDA
02198500	Savannah River near Clyo, Ga.	GEORGIA
12318500	Kootenai River near Copeland, Id.	IDAHO
13317000	Salmon River at Whitebird, Id.	IDAHO
05446500	Rock River near Joslin, Ill.	ILLINOIS
05599500	Big Muddy River at Murphysboro, Ill.	ILLINOIS
07022000	Mississippi River at Thebes, Ill.	ILLINOIS
03276500	Whitewater River at Brookville, Ind.	INDIANA
03374100	White River at Hazleton, Ind.	INDIANA
03378500	Wabash River at New Harmony, Ind.	INDIANA
06485500	Big Sious River at Akron, Ia.	IOWA
06486000	Missouri River at Sioux City, Ia.	IOWA
06877600	Smoky Hill River at Enterprise, Kans.	KANSAS
06892350	Kansas River at DeSota, Kans	KANSAS
07137500	Arkansas River near Coolidge, Kans.	KANSAS
07146500	Arkansas River at Arkansas City, Kans.	KANSAS
03215000	Big Sandy River at Louisa, Ky.	KENTUCKY
03216600	Ohio River at Greenup Dam near Greenup, Ky.	KENTUCKY
03254000	Licking River at Butler, Ky.	KENTUCKY
03277200	Ohio River at Markland Dam near Warsaw, Ky.	KENTUCKY
03290500	Kentucky River at Lock 2, at Lockport, Ky.	KENTUCKY
03303280	Ohio River at Cannelton Dam, Ky.	KENTUCKY
03321230	Green River near Beech Grove, Ky.	KENTUCKY
03438220	Cumberland River near Grand Rivers, Ky.	KENTUCKY
03609750	Tennessee River at Highway 60, near Paducah, Ky.	KENTUCKY
03612500	Ohio River at Dam 53 near Grand Chain, Ill.	KENTUCKY
02489500	Pearl River near Bogalusa, La.	LOUISIANA

Station No.	Station Name	State
02492000	Bogue Chitto near Bush, La.	LOUISIANA
07355500	Red River at Alexandria, La.	LOUISIANA
07369500	Tensas River at Tendal, La.	LOUISIANA
07373420	Mississippi River near St. Francisville, La.	LOUISIANA
07374525	Mississippi River at Belle Chasse, La.	LOUISIANA
07381600	Lower Atchafalaya River at Morgan City, La.	LOUISIANA
07385700	Bayou Teche at Keystone Lock, near St. Martinsville, La.	LOUISIANA
08015900	Calcasieu River near Lake Charles, La.	LOUISIANA
01017100	Aroostock River at Caribou, Me.	MAINE
01021050	St. Croix River at Milltown, Me.	MAINE
01491000	Choptank River near Greensboro, Md.	MARYLAND
01096550	Merrimack River above Lowell, Ma.	MASSACHUSETTS
01103500	Charles River at Dover, Ma.	MASSACHUSETTS
04045500	Tahquamenon River near Tahquamenon Paradise, Mich.	MICHIGAN
04108690	Kalamazoo River at Saugatuck, Mich.	MICHIGAN
04126520	Manistee River at Manistee, Mich.	MICHIGAN
04132052	Cheboygan River at Lincoln Ave. at Cheboygan, Mich.	MICHIGAN
04165500	Clinton River at Mount Clemens, Mich.	MICHIGAN
04165700	Detroit River at Detroit, Mich.	MICHIGAN
04024000	St. Louis River at Scanlon, Minn.	MINNESOTA
05112000	Roseau River below State Ditch 51 near Caribou, Minn.	MINNESOTA
05267000	Mississippi River near Royalton, Minn.	MINNESOTA
05330000	Minnesota River near Jordan, Minn.	MINNESOTA
05378500	Mississippi River at Winona, Minn.	MINNESOTA
07289000	Mississippi River at Vicksburg, Miss.	MISSISSIPPI
05490600	Des Moines River at St. Francisville, Mo.	MISSOURI
06818000	Missouri River at St. Joseph, Mo.	MISSOURI
06934500	Missouri River at Hermann, Mo.	MISSOURI
06054500	Missouri River at Toston, Mont.	MONTANA
06109500	Missouri River at Virgelle, Mont.	MONTANA
06130500	Musselshell River at Mosby, Mont.	MONTANA
06174500	Milk River at Nashua, Mont.	MONTANA
06185500	Missouri River near Culbertson, Mont.	MONTANA
06214500	Yellowstone River at Billings, Mont.	MONTANA
06308500	Tongue River at Miles City, Mont.	MONTANA
06329500	Yellowstone River near Sidney, Mont.	MONTANA
06686000	North Platte River at Lisco, Nebr.	NEBRASKA
06805500	Platte River at Louisville, Nebr.	NEBRASKA
10301500	Walker River near Wabuska, Nev.	NEVADA
10335000	Humboldt River near Rye Patch, Nev.	NEVADA
10351700	Truckee River near Nixon, Nev.	NEVADA
01404100	Raritan River near South Bound Brook, N.J.	NEW JERSEY
01463500	Delaware River at Trenton, N.J.	NEW JERSEY
07227140	Canadian River above N.Mex.-Tex. State Line, N. Mex.	NEW MEXICO
08313000	Rio Grande at Otowi Bridge, N. Mex.	NEW MEXICO
08358300	Rio Grande Conveyance Channel at San Marcial, N. Mex.	NEW MEXICO
08407500	Pecos River at Red Bluff, N. Mex.	NEW MEXICO
09368000	San Juan River at Shiprock, N. Mex.	NEW MEXICO
01358000	Hudson River at Green Island, N.Y.	NEW YORK
04260500	Black River at Watertown, N.Y.	NEW YORK
04295000	Richelieu River (Lower Champlain) at Rouses Point, N.Y.	NEW YORK
02080500	Roanoke River at Roanoke Rapids, N.C.	NORTH CAROLINA
02089500	Neuse River at Kinston, N.C.	NORTH CAROLINA
02129000	Pee Dee River near Rockingham, N.C.	NORTH CAROLINA
05124000	Souris River near Westhope, N. Dak.	NORTH DAKOTA
06337000	Little Missouri River near Watford City, N. Dak.	NORTH DAKOTA
03150000	Muskingum River at McConnelsville, Oh.	OHIO
03234500	Scioto River at Higby, Oh.	OHIO
03245500	Lower Miami River at Milford, Oh.	OHIO
03274600	Greater Miami River at New Baltimore, Oh.	OHIO
04208000	Cuyahoga River at Independence, Oh.	OHIO
07157950	Cimarror River near Buffalo, Okla.	OKLAHOMA
07161000	Cimarron River at Perkins, Okla.	OKLAHOMA
07178620	Verdigris River (Newt Graham Lock and Dam) near Inola, Okla.	OKLAHOMA
07231500	Canadian River at Calvin, Okla.	OKLAHOMA
07237500	North Canadian River at Woodward, Okla.	OKLAHOMA
07305000	North Fork Red River near Headrick, Okla.	OKLAHOMA
07331000	Washita River near Dickson, Okla.	OKLAHOMA
14048000	John Day River at McDonald Ferry, Oreg.	OREGON

Station No.	Station Name	State
14207500	Tualatin River at West Linn, Oreg.	OREGON
14301000	Nehalem River near Foss, Oreg.	OREGON
14372300	Rogue River near Agness, Oreg.	OREGON
01474500	Schuylkill River at Philadelphia, Pa.	PENNSYLVANIA
01540500	Susquehanna River at Danville, Pa.	PENNSYLVANIA
01553500	West Branch Susquehanna River at Lewisburg, Pa.	PENNSYLVANIA
01570500	Susquehanna River at Harrisburg, Pa.	PENNSYLVANIA
03049625	Allegheny River at New Kensington, Pa	PENNSYLVANIA
03085000	Monongahela River at Braddock, Pa.	PENNSYLVANIA
02136000	Black River at Kingstree, S.C.	SOUTH CAROLINA
02175000	Edisto River near Givhans, S.C.	SOUTH CAROLINA
06357800	Grand River at Little Eagle, S.D.	SOUTH DAKOTA
06438000	Belle Fourche River near Elm Springs, S.D.	SOUTH DAKOTA
06439300	Cheyenne River at Cherry Creek, S.D.	SOUTH DAKOTA
06452000	White River near Oacoma, S.D.	SOUTH DAKOTA
06478500	James River near Scotland, S.D.	SOUTH DAKOTA
03571850	Tennessee River at South Pittsburg, Tenn.	TENNESSEE
07032000	Mississippi River at Memphis, Tenn.	TENNESSEE
07228000	Canadian River near Canadian, Tex.	TEXAS
07308500	Red River near Burkburnett, Tex.	TEXAS
08030500	Sabine River near Ruliff, Tex.	TEXAS
08041000	Neches River at Evadale, Tex.	TEXAS
08066500	Trinity River at Romayor, Tex.	TEXAS
08068000	West Fork San Jacinto River near Conroe, Tex.	TEXAS
08082000	Salt Fork Brazos River near Aspermont, Tex.	TEXAS
08098290	Brazos River near Highbank, Tex.	TEXAS
08116650	Brazos River near Rosharon, Tex.	TEXAS
08162000	Colorado River at Wharton, Tex.	TEXAS
08176500	Guadalupe River at Victoria, Tex.	TEXAS
08188500	San Antonio River at Goliad, Tex.	TEXAS
08210000	Nueces River near Three Rivers, Tex.	TEXAS
08212400	Los Olmos Creek near Falfurrias, Tex.	TEXAS
08377200	Rio Grande at Foster Ranch near Langtry, Tex.	TEXAS
08447410	Pecos River near Langtry, Tex.	TEXAS
08459000	Rio Grande at Laredo, Tex.	TEXAS
08475000	Rio Grande near Brownsville, Tex.	TEXAS
09180500	Colorado River near Cisco, Utah	UTAH
09315000	Green River at Green River, Utah	UTAH
09379500	San Juan River near Bluff, Utah	UTAH
10126000	Bear River near Corinne, Utah	UTAH
10141000	Weber River near Plain City, Utah	UTAH
10224000	Sevier River near Lynndyl, Utah	UTAH
01673000	Pamunkey River near Hanover, Va.	VIRGINIA
02035000	James River at Cartersville, Va.	VIRGINIA
02049500	Blackwater River near Franklin, Va.	VIRGINIA
12045500	Elwha River at McDonald Branch near Port Angeles, Wash.	WASHINGTON
12433000	Spokane River at Long Lake, Wash.	WASHINGTON
12510500	Yakima River at Kiona, Wash.	WASHINGTON
03201300	Kanawha River at Winfield, W. Va.	WEST VIRGINIA
04085000	Fox River at Wrightstown, Wis.	WISCONSIN
05340500	St. Croix River at St. Croix Falls, Wis.	WISCONSIN
05369500	Chippewa River at Durand, Wis.	WISCONSIN
05407000	Wisconsin River at Muscoda, Wis.	WISCONSIN

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