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# Trends in pesticide concentrations and use for major rivers of the United States



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## HIGHLIGHTS

- · Concentration and use trends were assessed for 11 pesticides in 38 US rivers.
- · Concentration and use trends mostly agreed for agricultural pesticides.
- Regulations and urban-stream trends explain trends related to nonagricultural use.
- · For most trend discrepancies, concentration increased more than use.
- Unaccounted use may contribute to greater concentration increases in some cases.

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### ABSTRACT

Trends in pesticide concentrations in 38 major rivers of the United States were evaluated in relation to use trends for 11 commonly occurring pesticide compounds. Pesticides monitored in water were analyzed for trends in concentration in three overlapping periods, 1992–2001, 1997–2006, and 2001–2010 to facilitate comparisons among sites with variable sample distributions over time and among pesticides with changes in use during different periods and durations. Concentration trends were analyzed using the SEAWAVE-Q model, which incorporates intra-annual variability in concentration and measures of long-term, mid-term, and short-term streamflow variability. Trends in agricultural use within each of the river basins were determined using interval-censored regression with high and low estimates of use.

Pesticides strongly dominated by agricultural use (cyanazine, alachlor, atrazine and its degradate deethylatrazine, metolachlor, and carbofuran) had widespread agreement between concentration trends and use trends. Pesticides with substantial use in both agricultural and nonagricultural applications (simazine, chlorpyrifos, malathion, diazinon, and carbaryl) had concentration trends that were mostly explained by a combination of agricultural-use trends, regulatory changes, and urban use changes inferred from concentration trends in urban streams. When there were differences, concentration trends usually were greater than use trends (increased more or decreased less). These differences may occur because of such factors as unaccounted pesticide uses, delayed transport to the river through groundwater, greater uncertainty in the use data, or unquantified land use and management practice changes.

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#### 1. Introduction

The use of pesticides has a range of benefits, including increased food production and reduction of insect-borne diseases, but also raises concerns about possible adverse effects on the environment, including water quality. Once released into the environment, pesticides can move through the hydrologic system to streams and groundwater, where they may have unintended effects on humans, aquatic life, or wildlife. Understanding the long-term trends of pesticide concentrations in the hydrologic system is essential to understanding their potential for adverse effects, how past use has affected concentrations in streams and rivers, and how future changes in use or management may affect concentration trends.

Previous analyses of concentration trends in rivers and streams of the United States (US) Corn Belt showed that trends in major rivers and their tributaries were largely consistent with each other and with use trends, and that the concentration trends in large rivers provide a smoothed indication of large scale trends (Sullivan et al., 2009; Vecchia et al., 2009). Use data generally are not available for estimation of nonagricultural uses of pesticides, but an analysis of concentration trends in urban streams showed varying patterns in trend direction

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depending on analysis period, region of the US, chemical, and regulatory actions (Ryberg et al., 2010). Stone et al. (2014) summarized trends for a subset of pesticides used in agricultural and urban settings over the last two decades (1992–2011) and reported widespread trends in pesticide concentrations in surface water that varied in direction in response to changes in use driven by regulatory actions and new pesticide introductions.

This paper presents an analysis of trends in pesticide concentrations and agricultural-use intensity (agricultural use) for 11 compounds in 38 major rivers of the US (Fig. 1, Table 1), a subset of the trend analysis of Ryberg et al. (2014). The pesticides include the herbicides cyanazine, alachlor, atrazine and its degradate deethylatrazine (DEA), metolachlor, and simazine; and the insecticides chlorpyrifos, malathion, diazinon, carbofuran, and carbaryl. All 11 compounds are among the top 20 most frequently detected in US streams and rivers (based on those analyzed by the U.S. Geological Survey National Water-Quality Assessment Program; Stone et al., 2014). The analysis was limited to pesticides that met the specific data requirements for trend analysis and data deficiencies leave out many important compounds, such as glyphosate, pyrethroids, and neonicotinoids. Glyphosate, for example, is "difficult and costly to measure" and assessment efforts in the US have been "limited primarily to regional, targeted, or short-term studies" (Stone et al., 2014). Supplementary Table 1 contains chemical properties affecting the transport and fate of the compounds.

The analysis of paired concentration and use trends in the present study contributes to a better understanding of how long-term trends in concentration are affected by use and regulatory changes. The compounds included have a wide variety of uses and the major rivers evaluated are distributed across the US. National annual agricultural use estimates for the five herbicides and five insecticides are shown in Fig. 2. The use estimates are shown in terms of the types of crops they are used on and the estimates highlight changes in pesticide use and regulation and changes in national cropping patterns. Supplementary Table 2 contains additional information about the pesticides, including their nonagricultural uses. The online version of this article includes an interactive map of the sites as supplementary geospatial information.

Pesticide concentration trends in these major rivers potentially reflect various combinations of large-scale changes in pesticide use (such as those due to crop changes, regulatory changes, or market forces), changes in land use (such as increased urbanization), changes in management practices (such as tillage practices, tile drainage, or conservation buffer strips), changes in climatic conditions, and other factors individually or in combinations that were prevalent in their respective regions. Generally, trends were only assessable for pesticides that were used extensively, are relatively water soluble, or are persistent enough to be frequently detected in filtered water at sampling sites, because these conditions result in sufficient detections for trend analysis.

The major contribution of this study compared to the previous Corn Belt and urban pesticide trend studies is that this study incorporates recently compiled agricultural use data for the compounds and compares and contrasts the concentration and use trends for major rivers distributed throughout the US. By identifying the directions, magnitudes, and statistical significance of trends, in context with changes in

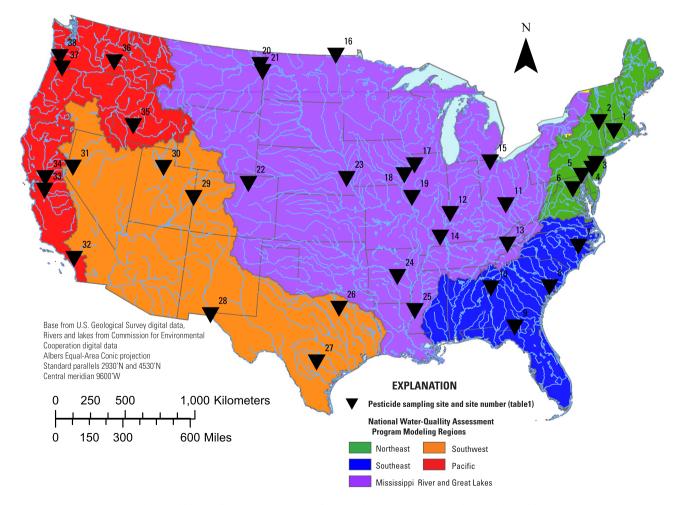


Fig. 1. Pesticide sampling sites on major rivers of the United States. Sites are described by number in Table 1.

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