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Imidacloprid in United States Rivers, 2013−2022: Persistent Presence and Emerging Chronic Hazard

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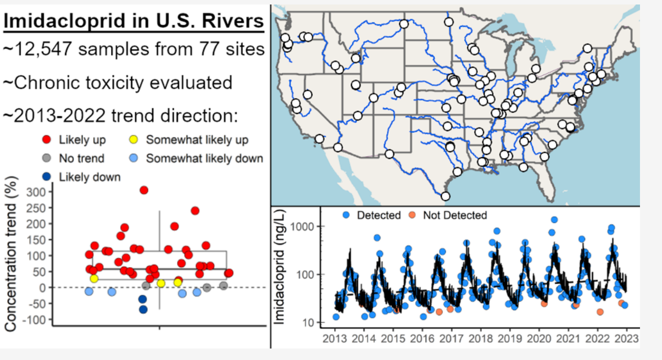
ABSTRACT: Imidacloprid, a neonicotinoid insecticide, is used for agricultural and nonagricultural purposes and is toxic to nontarget organisms at low concentrations in aquatic ecosystems. A total of 12,547 water samples were collected from 2013 to 2022 from 77 rivers across the United States (U.S.) and were analyzed to evaluate detections and temporal trends in imidacloprid concentrations. Imidacloprid was detected in 44% of all samples, and the mean concentration, adjusted for nondetect samples, of 24.9 ng/L (median = 11.9 ng/L) was more than twice the chronic benchmark for freshwater invertebrates (10 ng/L). This potential hazard to aquatic life was persistent, with 44% of the sites having a median concentration exceeding the chronic benchmark. Half of the sites (n = 38) had increasing trends, including large river sites along the Mississippi River. The mean increase was 10.6 ng/L over the past decade, while only six sites indicated decreasing trends. The estimated total loading of imidacloprid delivered to the Gulf of America from 2013 to 2022 was 129,489 kg (142.7 U.S. ton). The extensive presence of imidacloprid in U.S. waterways, the high percentage of sites with trends of increasing concentrations, and the prevalence of concentrations exceeding chronic benchmarks suggest widespread persistent risks to ecosystem health.



KEYWORDS: *imidacloprid, pesticide, neonicotinoid, contamination, toxic, water quality, trend, river*

■ INTRODUCTION

Pesticides are chemicals used for both agricultural and nonagricultural purposes and are contaminants of global concern having unintended consequences to nontarget organisms[1] that pose potential threats to both human[2,3] and ecosystem health.[4−8] Due to their powerful nerve-damaging effects as systemic neurotoxins, neonicotinoids are the most widely used class of insecticides in the world for treating a wide variety of sucking and chewing insects.[9] In 2014, neonicotinoids accounted for >25% of global insecticide sales,[10] of which thiamethoxam, clothianidin, and imidacloprid accounted for almost 85% of total neonicotinoid sales. The global market value of neonicotinoids in 2018 was $4.752 billion and accounted for the largest percentage of the global market share for insecticides (24%).[11]

Imidacloprid, one of the most widely used insecticides in North America, provides effective pest control for a variety of agricultural and nonagricultural uses.[12] Imidacloprid is commonly used as a broad-spectrum, prophylactic seed treatment for major agricultural crops.[13,14] Because imidacloprid has high solubility (610 mg/L at 20 °C) and moderate mobility (soil water partition coefficient normalized to soil organic carbon content [K_{oc}] = 266 L/kg), imidacloprid poses substantial concerns regarding leaching from seed treatments into surrounding soils, surface runoff into streams,[12] and subsurface infiltration leading to groundwater transport and discharge into surface waters,[15,16] including via subsurface tile drains.[17] Imidacloprid is also used in developed areas for nonagricultural pest control such as termite deterrent,[12] invasive species control in protected areas,[18] application on lawns, ornamental plants, commercial landscaping, and turfgrass,[19] forestry,[20] and applied as spot-on parasiticides to companion animals.[21] Due to the large number of uses in developed areas, imidacloprid has frequently been detected in wastewater.[21−24] The widespread use of imidacloprid in the United States has led to frequent detections in groundwater and streams draining both agricultural and developed lands,[23,25−31] as well as in drinking water.[2,3,32]

Imidacloprid is highly toxic to sensitive species, particularly insects, at low concentrations.[33−36] The U.S. Environmental Protection Agency (USEPA) aquatic life chronic and acute

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benchmarks for freshwater invertebrates in 2025 were 10 and 385 ng/L, respectively.[37] Previous studies have also observed sublethal effects from imidacloprid on vertebrate wildlife including fish, amphibians, reptiles, birds, and mammals.[38] The USEPA determined that imidacloprid is likely to adversely affect 79% of endangered or threatened species in the U.S. and 83% of critical habitats.[12] Imidacloprid has been found to be a major contributor to potential ecotoxicity of aquatic macroinvertebrates in Midwestern U.S. streams[6] as well as in national assessments.[4,5,39−41]

In a previous study of U.S. rivers from 2013 to 2017, imidacloprid was the most frequently detected insecticide and posed the greatest potential threat to aquatic life, exceeding benchmark concentrations at 60 of the 74 sites.[40] The analysis presented here provides an update to the 2013−2017 study to determine if concentrations exceeding benchmarks were persistent through 2022 and to quantify potential temporal changes over a decade of imidacloprid sampling. To investigate the occurrence and potential effects of imidacloprid in rivers in the United States, data collected from 2013 to 2022 at 77 sites spanning different dominant land use categories, drainage areas, and geographic regions were analyzed. The goals of this study were to (1) characterize the occurrence of imidacloprid from a national network of rivers and streams and quantify the potential ecological risk by comparing measured concentrations to relevant aquatic life benchmarks, and (2) assess trends in imidacloprid concentrations and loads from 2013 to 2022 using a model that accounts for seasonal- and flow-related variability of pesticide concentrations (SEAWAVE-Q).[42] Understanding how concentrations and loads vary over time is necessary to document temporal changes and identify potential threats to aquatic ecosystems.

MATERIALS AND METHODS

Study Area. The U.S. Geological Survey (USGS) National Water Quality Network (NWQN) was established to enable a national-scale understanding of surface water quality conditions from a range of land use settings, stream sizes, and geographic regions.[43] Previous pesticide trend studies occurred prior to widespread imidacloprid monitoring.[44−48] The USGS began routinely monitoring imidacloprid, using a newly developed analytical method, in 2013;[49] therefore, this is the first study that has a decade of samples to analyze annual trends in concentration and load. All sites included in the NWQN are collocated at streamgages, allowing for the computation of constituent loads. Streamflow and water quality data from this network were collected and analyzed using consistent methods to facilitate spatial and temporal analyses of water quality at all sites. Imidacloprid samples from 77 sites in the NWQN were collected 12 to 24 times per year from 2013 to 2022 during a range of streamflow conditions.[50]

Study sites were categorized by dominant land use, drainage area, and geographic region to evaluate detection frequencies, concentration ranges, and benchmark exceedances across different landscapes following previously defined categories[51] that utilized a 60 m resolution national land use grid.[52] Drainage area of the sites ranged from 41 to 2,938,531 km^2 and was divided into three categories: small (<10,000 km^2), medium (10,000−100,000 km^2), and large (>100,000 km^2). Most of the drainage areas were large enough that the surface water quality was affected by more than one land use category. Sites, however, were categorized by dominant land use (i.e., agriculture, developed, mixed, or undeveloped).[40] The exact formula used to define the dominant land use categories can be found in a published data release.[51] States with similar crop production patterns were combined to categorize five broad geographic regions of the United States (Northeast, South, Midwest, West, or Pacific).

Data Compilation. Imidacloprid concentration data (n = 13,058) from 77 NWQN sites sampled from January 1, 2013 to December 31, 2022 were downloaded from the Water Quality Portal (https://www.waterqualitydata.us/) using the R package "dataRetrieval".[50,53,54] Samples with a reported detected concentration of 0 (n = 35) were excluded as well as samples with raised reporting levels greater than 25 ng/L (n = 346), which was near the median concentration of detected samples (27.3 ng/L). Weekly sampling was deemed the highest appropriate frequency for trend analysis.[55] Therefore, all samples from a site were categorized by calendar weeks (from Sunday to Saturday), and if multiple samples were collected within the same week, the sample collected nearest to 12 p.m. on Wednesday was kept for trend analysis. This screening resulted in the exclusion of 130 samples. The remaining 12,547 samples were used for all analyses.

This study used water samples analyzed by the USGS National Water Quality Laboratory using the direct aqueous injection liquid chromatography−tandem mass spectrometry method (laboratory schedules 2437 and 2447),[56] in which filtration occurred prior to sample injection. Field blank (n = 313), laboratory blank (n = 2,005), replicate (n = 572), laboratory reagent spike (n = 1,170), and field matrix spike (n = 449) quality assurance samples collected for this method indicated the results from environmental samples met data quality objectives.[57−60] Only one field blank sample had a detection (9.3 ng/L at USGS site 01184000 on 7/22/2020) and no detections were observed from laboratory blank samples.[57] The median percent difference from 572 replicate samples was 14.0%.[57,59] In 1,170 laboratory reagent spike samples, the median percent recovery was 97.2%, and 96.9% of samples had a recovery between 70% and 130%.[57,58,60] The median recovery from 449 surface water field matrix spike samples, however, was 131%, and while 48.8% of samples had a recovery between 70% and 130% (57,58,60), 88.0% of the field matrix spike samples had a recovery greater than 100%, which indicates a positive matrix bias.

Most mean daily streamflow data during the analysis period were previously published.[61] Streamflow data from October 1, 2022 to December 31, 2022 were downloaded from the USGS National Water Information System using the R package "dataRetrieval"[54] for the majority of study sites. Data for one site (Mississippi River above Vicksburg at Mile 438, Mississippi [USGS site 322023090544500], hereafter Mississippi River above Vicksburg) were computed as the difference in streamflow from two adjacent locations,[61] and data from two sites (Mississippi River near St. Francisville, Louisiana [USGS site 07373420], hereafter Mississippi River near St. Francisville, and (Corps of Engineers) Atchafalaya River at Melville, Louisiana [USGS site 07381495], hereafter Atchafalaya River at Melville) were obtained from external sources described by Lee.[61] A complete streamflow record was required to compute imidacloprid loads; therefore, the R package "waterData" was used to fill small gaps of missing data,[62] representing less than 0.3% of total observations. The imidacloprid load delivered to the Gulf of America (formerly Gulf of Mexico) from two sites (Mississippi River near St. Francisville and Atchafalaya River at Melville) was compared

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to estimated county-level imidacloprid agricultural use within their respective watersheds.[63] Basin boundaries for each site were obtained,[54] merged together,[64] and clipped to county boundaries[65] to identify intersecting counties within the combined basin boundary.

Data Analysis. Imidacloprid detection frequency was determined by dividing the number of detected samples by the total number of samples at individual sites, categories of sites based on dominant land use, drainage area, and geographic region, as well as across the entire data set. The detection frequency during the first half of the study period (2013−2017) was compared to the second half of the study period (2018−2022) using a two-sided test of equal proportions with the R package "stats",[53] where significant differences were determined when $p < 0.05$. The percent of samples exceeding the chronic (10 ng/L) and acute (385 ng/L) freshwater invertebrate aquatic life benchmarks[37] were also calculated. Differences in concentrations among dominant land use categories, drainage areas, and geographic regions were evaluated with Tukey's honestly significant difference test using the R package "agricolae",[66] where significant differences were determined when $p < 0.05$ using log-transformed concentration results with nondetect sample results replaced with one-half of the detection limit.[67] Detection frequency and concentrations were analyzed for seasonal and annual patterns among the entire data set, categories of sites, and individual sites. Seasonality represented by annual variability in detected samples was calculated by dividing the difference in the maximum and minimum monthly median concentrations by the minimum monthly median concentration and multiplying by 100 to quantify the extreme values as a percentage of the lowest month's concentration for all sites, individual sites, and categories of sites (eq 1):

$$\text{seasonality (\%)} = \left(\frac{C_{\text{max}} - C_{\text{min}}}{C_{\text{min}}}\right) \times 100 \tag{1}$$

where C_{max} is the maximum of the monthly median concentration and C_{min} is the minimum of the monthly median concentration. Imidacloprid concentration−discharge slopes were calculated by fitting a linear regression to the log-transformed concentration and streamflow data.

Trends in imidacloprid concentrations were assessed using the statistical model Seasonal Wave (SEAWAVE) and adjustment for streamflow (Q; SEAWAVE-Q) from the R package "seawaveQ".[42,68] The SEAWAVE-Q model is a parametric regression model that uses a dimensionless seasonal wave and is designed to analyze seasonal- and flow-related variability and trends in pesticide concentrations.[42] This trend method uses a parametric survival regression model using maximum likelihood methods for censored data. A pulse input function is selected in seawaveQ from 14 options based on the distinct application season and the model half-life from one to four months, creating a total of 56 available choices for the seasonal wave function.[42] The observed concentration data are used to select the best wave function for each model. The model was specifically designed to assess pesticide trends and caters to data characteristics that are common with pesticide data, including a high degree of censoring and intermittent or changing sampling frequencies over the trend period. Likewise, SEAWAVE-Q is designed to assess data with complex relations between streamflow and concentration and strong seasonality, making it an ideal choice for pesticide data.

Sample size criteria used to determine applicability of the SEAWAVE-Q model at each NWQN site followed previous established recommendations[55] including at least 10 detected values, at least five years of samples including at least six samples in at least two of the first and last five years of the study, and a daily streamflow record. In addition, a minimum detection frequency of 20% was used to select sites for trend analysis, a recommended threshold for robust statistical analysis.[69,70] Forty-seven of the 77 sites met these criteria,[50] representing all geographic regions and drainage area categories but primarily agriculture, developed, or mixed dominant land use categories. Short-term (1 day), medium-term (30 day), and long-term (1 year) streamflow anomalies were calculated using the R package "waterData"[62] to help account for streamflow variability and aid in the prediction of daily pesticide concentrations.[71] The fifth percentile, median, and ninety-fifth percentile concentrations were estimated for the entire data set, categories of sites, and individual sites that were selected for trend analysis while accounting for nondetect samples using the regression on order statistics method[70] by employing the R package "NADA".[72] The mean concentration was estimated using the Kaplan−Meier method from the R package "NADA'.[72] All references to concentration summary statistics refer to those adjusted for nondetect samples unless otherwise stated.

Trend results were reported in both concentration units (ng/L) and percent change over the study period and interpreted as the trend in the flow-adjusted annual median concentration.[73] The annual load trend from SEAWAVE-Q cannot be different from the trend in the flow-adjusted annual median concentration when there is no trend in flow. A likelihood-based approach using the two-sided p-value associated with the significance level of the trend was used to report trend results.[74] A trend was considered "likely up" or "likely down" when the trend likelihood value was between 0.85 and 1.0, while a trend was considered "somewhat likely up" or "somewhat likely down" when the likelihood value was greater than 0.70 and less than or equal to 0.85. When the likelihood value was less than or equal to 0.70, the trend was considered "about as likely as not". Daily imidacloprid concentration estimates from seawaveQ were corrected for retransformation bias based on the quasi-maximum likelihood estimator[75] following eq 3 in the work by Ryberg and York[42] and then multiplied by mean daily streamflow to compute the daily load. Daily loads of imidacloprid at each site were summarized by year to determine the annual load. Complete SEAWAVE-Q model results are published in a data release[50] including the model parameter outputs, concentration trend results, and annual load calculations.

To assess how changes in laboratory performance over time may influence the interpretation of trend results, the analytical recovery was calculated. Concentrations from environmental samples were corrected using the set of 1,170 laboratory reagent spikes.[57,58,60] Using a previously published approach,[76] a lowess model was fit to percent recovery for all laboratory reagent spike samples from 2013 to 2022 (Figure S1). Individual sites showed variability in field matrix spike recovery over time; however, sampling frequency did not permit the evaluation of site-specific field matrix recovery corrections. Recovery was estimated for each day during the study period from the lowess model, based on the laboratory reagent spike samples, and was matched to each sample collection date and used to correct the measured concentrations to 100% recovery

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Table 1. Summary[a] of Imidacloprid Detections and Concentrations in 77 U.S. Rivers from 2013 to 2022 Relative to Aquatic Life Acute and Chronic Benchmark Standards for Freshwater Invertebrates[37]

Category	Sites (*n*)	Samples (*n*)	Detection (%)	Chronic Benchmark Exceedance (%)	Acute Benchmark Exceedance (%)	Median (ng/L)	Mean (ng/L)	Season-ality (%)
All sites	77	12,547	43.7	38.9	0.3	11.9	24.9	93
Trend sites	47	8,043	64.2	58.4	0.5	16.8	34.0	111
Agriculture	33	5,463	49.9	44.3	0.4	12.8	28.9	148
Developed	13	2,664	75.2	71.9	0.6	23.9	40.1	73
Mixed	17	2,358	22.2	16.5	0.0	8.1	11.8	289
Undeveloped	14	2,062	11.2	7.7	0.0	6.9	8.7	148
Small	27	5,540	53.4	49.7	0.6	14.6	31.2	95
Medium	27	4,031	39.4	34.4	0.2	11.4	24.5	126
Large	23	2,976	31.4	25.0	0.0	9.4	13.6	226
Northeast	9	1,563	31.9	26.8	0.7	9.6	25.3	191
South	20	3,322	69.7	49.7	0.3	21.8	38.5	112
Midwest	25	3,864	45.1	38.4	0.4	11.5	24.2	170
West	14	2,203	16.7	13.4	0.0	7.9	10.7	66
Pacific	9	1,595	34.9	31.7	0.1	10.4	17.0	107

[a]Imidacloprid sample results summary for all sites, trend sites, and various categories of sites; the number of sites and samples per category, the percent of samples with a detected concentration, and the percent of samples exceeding the chronic and acute benchmark concentrations; the median and mean concentration adjusted for nondetect samples using the regression on order statistics and Kaplan−Meier methods,[72] respectively; and the seasonality, in percent increase, calculated by dividing the difference in the maximum and minimum monthly median concentrations by the minimum monthly median concentration and multiplying by 100.

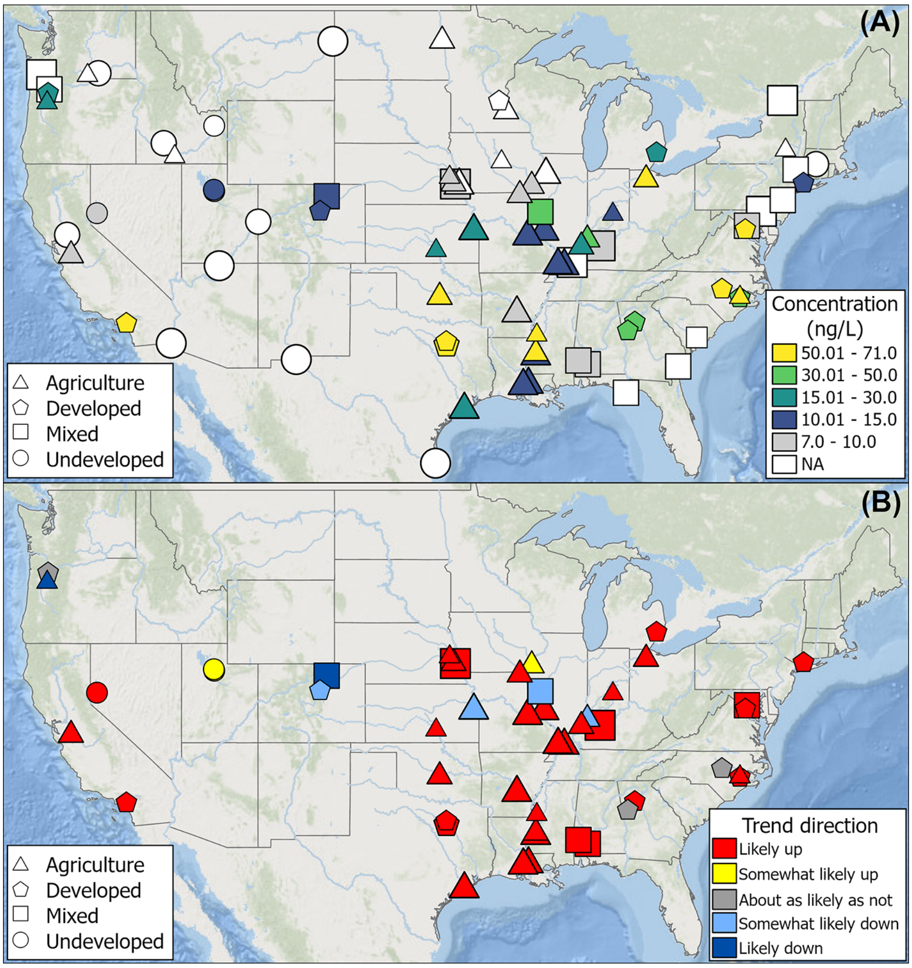

Figure 1. Location of 77 sites sampled in this study colored by (A) median imidacloprid concentration in ng/L and (B) trend direction. The median concentration was adjusted for nondetects using the regression on order statistics method.[72] White symbols in (A) indicate sites with a detection frequency below 20% in which trend analysis and median concentration calculation were not conducted (NA) and thus are not shown in (B). Symbol size is proportional to drainage area size class.[50] Base map sources include Esri, Garmin, GEBCO, and NaturalVue.

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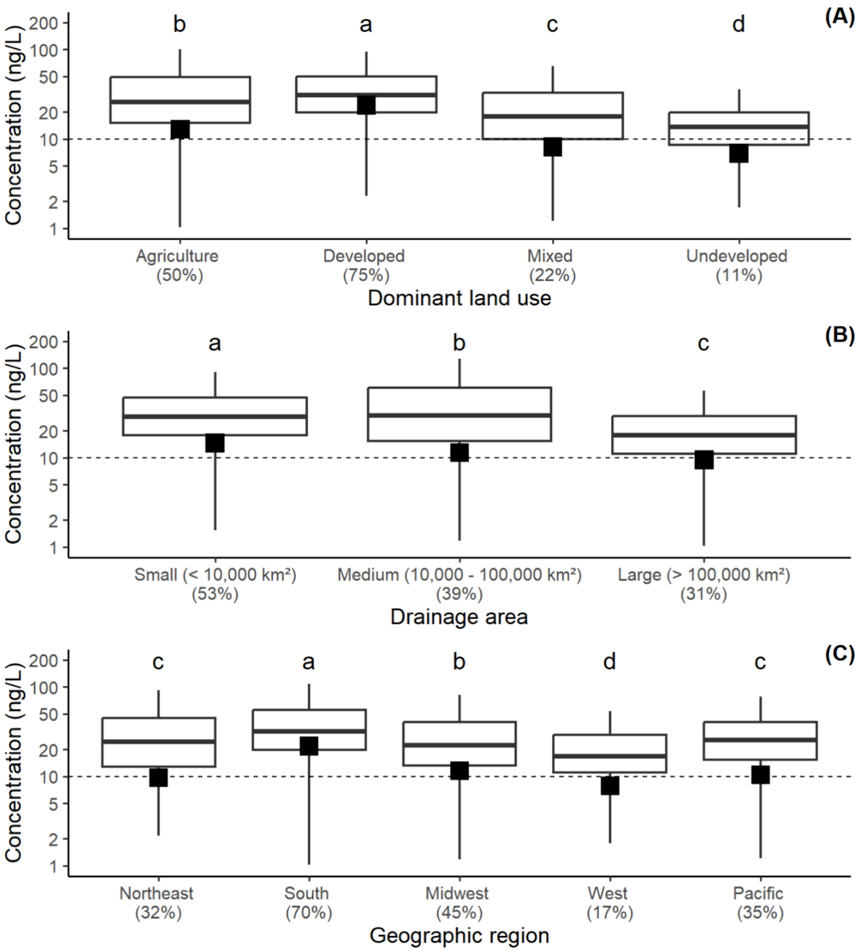

Figure 2. Imidacloprid concentrations from all 77 sites sampled in this study from 2013 to 2022 summarized by (A) land use, (B) drainage area, and (C) region. The box represents the interquartile range of detected sample concentrations with the median denoted with the middle line. The lower and upper lines extend 1.5 times the interquartile range from the top and bottom of the box. Percentages in the *x*-axis labels indicate the detection frequency. Large squares indicate the median monthly concentration adjusted for nondetect samples using the regression on order statistics method.[72] The horizontal dashed line denotes the U.S. Environmental Protection Agency chronic (10 ng/L) aquatic life benchmark for freshwater invertebrates.[37] Different lowercase letters above the boxplots denote significant differences according to Tukey's honestly significant difference test at the 0.05 significance level.[66]

from environmental samples by dividing the uncorrected measured concentration by the modeled recovery (eq 4 from Martin et al.).[76] Nondetects were not corrected for recovery. Trend results are presented from the recovery-corrected data, but both the uncorrected as well as the recovery-corrected trend results are published for comparison.[50]

RESULTS AND DISCUSSION

Imidacloprid: A Widespread and Persistent Hazard. Imidacloprid was detected at most sites (44% of 12,547 samples; 72 out of 77 sites; Table 1) with the detection frequency significantly increasing from 42.0% in the first half of the study (2013−2017; total samples = 6,467) to 45.4% in the second half (2018−2022; total samples = 6,080).[50] The mean imidacloprid concentration adjusted for nondetects of 24.9 ng/L (median = 11.9 ng/L) was more than twice the USEPA aquatic life chronic benchmark for freshwater invertebrates. Chronic benchmark exceedances were frequent (39%) and widespread, with most sites observed to have at least one measured concentration that exceeded the chronic benchmark (88%). In comparison, far fewer samples (0.3%) and sites (12%) exceeded the acute benchmark. This chronic hazard to aquatic life was persistent with 44% of the sites having a median concentration exceeding the chronic benchmark.[50] While land use, drainage area, and geographic region (Figures 1 and 2; Table 1) influenced detections and benchmark exceedances, hazard to aquatic life due to chronic exposure to imidacloprid occurred throughout the nation, indicating that imidacloprid is a pervasive and widespread potential threat to ecosystem health across the United States.

Land use is important to consider when characterizing pesticide frequency of detection and concentration. Developed and agricultural watersheds generally had higher detection frequencies, concentrations, and more frequent chronic benchmark exceedances of imidacloprid compared to mixed or undeveloped land use (Figure 2; Table 1). As previously documented,[28] imidacloprid concentrations in developed watersheds averaged (mean = 40.1 ng/L; median = 23.9 ng/L) above those in agricultural watersheds (mean = 28.9 ng/L; median = 12.8 ng/L); thus, the frequency at which imidacloprid concentrations in samples exceeded the chronic benchmark in developed watersheds was nearly twice that observed in agricultural watersheds (developed = 71.9%; agricultural = 44.3%). Mean imidacloprid concentrations from

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other land uses such as mixed and undeveloped watersheds were slightly above and below the chronic benchmark (11.8 and 8.7 ng/L, respectively).

Watershed size and geographic region are also important context to understand imidacloprid risks to nontarget species. Detection and chronic exceedance frequencies of imidacloprid were greater in small watersheds (53.4% and 49.7%, respectively; <10,000 km^2), followed by medium watersheds (39.4% and 34.4%, respectively; 10,000−100,000 km^2) and large watersheds (31.4% and 25.0%, respectively; >100,000 km^2). However, the mean concentrations of imidacloprid for small (31.2 ng/L; median = 14.6 ng/L), medium (24.5 ng/L; median = 11.4 ng/L), and large (13.6 ng/L; median = 9.4 ng/L) watersheds all exceeded the chronic benchmark. Small watersheds had significantly greater imidacloprid concentrations compared to medium and large watersheds, with large watersheds having the lowest concentrations, a finding documented in other studies.[77,78] The higher concentrations observed in small watersheds may be attributed to a greater proportion of agricultural or developed land use, which often have increased pesticide application and reduced dilution capacity compared to larger watersheds.[77] Ten sites had chronic benchmark exceedance frequencies of imidacloprid greater than 90%, all of which were small or medium developed or agricultural watersheds primarily located in the South region.[50]

Imidacloprid concentrations in U.S. rivers were significantly greater in the South region (mean = 38.5 ng/L; median = 21.8 ng/L), followed by the Midwest (24.2 ng/L; 11.5 ng/L), Northeast (25.3 ng/L; 9.6 ng/L), Pacific (17.0 ng/L; 10.4 ng/L), and West (10.7 ng/L; 7.9 ng/L) regions (Figure 2; Table 1). Thirty sites had fewer than 40 detections (less than 20% detection frequency) during the study period; therefore, a robust trend analysis could not be conducted.[50] Most of these sites were medium or large sized watersheds with undeveloped or mixed land use (Figure 1). Estimated agricultural use of imidacloprid for row crops (e.g., corn, soybeans, wheat, and cotton), orchards, and vegetables varies throughout the United States but is relatively higher in the South and Midwest regions relative to other parts of the study area,[63] which may partially explain the observed geographic patterns. In the South study region, corn, soybeans, and cotton are the dominant crops, and the abundance of cotton and soybean production was linked to neonicotinoid resistance of an insect herbivore (*Frankliniella fusca*) that has led to a large increase in insecticide use on cotton.[79] Neonicotinoids used as cotton seed treatment were detected in soil for months post planting and present in some wetlands.[80] In the Midwest study region, corn and soybeans are the dominant crops in which seeds are typically treated with neonicotinoids including imidacloprid, clothianidin, and thiamethoxam.[81] Research in this region has detected the widespread occurrence of these neonicotinoids in surface water,[30] groundwater,[16,27,31,82] wetlands,[83−86] drinking water,[2] sewage wastewater,[23,24] and wastewater from ethanol production,[87,88] with imidacloprid concentrations and detections often lower than those of both thiamethoxam and clothianidin.[30] Study sites in the Northeast region had primarily developed or mixed dominant land uses, with developed sites indicating the highest concentrations of imidacloprid, which presents challenges for assessing the relation to estimated pesticide use since no accurate information on the nonagricultural use of pesticides is available.[63] However, previous studies have demonstrated consistent detections of imidacloprid, with concentrations typically exceeding chronic thresholds for freshwater macroinvertebrates, in urban streams throughout the United States.[23,26,29,41,89−93] The sites with the highest concentrations in the South study region had predominantly developed land use, which partially contributed to this region showing higher concentrations compared to the other regions.

The greatest risks to aquatic life from exposure to imidacloprid were observed in small to medium sized agricultural or developed watersheds. Acute benchmark exceedances were rare among all samples, but all of them occurred in small or medium agricultural or developed watersheds with two sites (Accotink Creek near Annandale, Virginia [USGS site 01654000]; small, developed, Northeast; 4.9% acute benchmark exceedance; Bogue Phalia near Leland, Mississippi [USGS site 07288650], hereafter Bogue Phalia near Leland; small, agricultural, South; 3.5% acute benchmark exceedance) accounting for half of the acute exceedances (Figure 3). Larger watersheds often have less variable pesticide

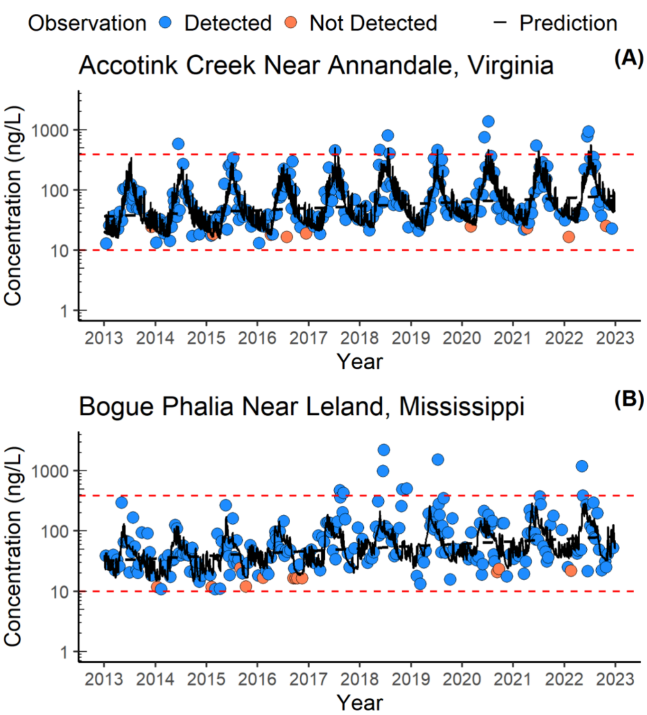

Figure 3. Observed imidacloprid concentrations and daily predicted concentration at study sites in two rivers in the United States: (A) Accotink Creek near Annandale, Virginia (USGS site 01654000) and (B) Bogue Phalia near Leland, Mississippi (USGS site 07288650). Detected samples are shown as blue circles, while nondetect samples are orange. The solid black line represents the daily predicted concentration from seawaveQ, and the dashed black line shows the trend in concentration. The two red dashed lines indicate the chronic (10 ng/L) and acute (385 ng/L) aquatic life benchmarks for freshwater invertebrates.[37]

concentrations compared to their component tributaries due to the timing of the delivery of water to the mainstem from the tributaries[78] as well as increased capacity for dilution from undeveloped land use associated with smaller pesticide application rates.[77] As a result, small watersheds tend to show more rapid fluctuations in pesticide concentrations and higher peak concentrations following storm events compared to larger watersheds; however, larger watersheds may be more

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likely to record moderate pesticide concentrations for extended periods due to longer duration runoff chemographs following storm events.[78] Nine large watersheds had a median imidacloprid concentration that exceeded the chronic benchmark, including the outlet of the Mississippi River (Mississippi River near St. Francisville; 12.1 ng/L; detection frequency = 69.3%; chronic exceedance frequency = 51.3%).

Imidacloprid detections often showed a seasonal pattern whereby the greatest concentrations and detection frequencies generally occurred in the late spring and summer months (Figure 4), which may be related to seasonal application rates;[94] however, seasonal variability ranged across the study area by land use, drainage area, and region. For example, seasonality was greater in agricultural watersheds compared to developed ones (Figure S2; Table 1), a finding documented before,[26,28] but similar to undeveloped watersheds and less than mixed use watersheds. Seasonality was more pronounced in large watersheds, followed by medium and small ones (Figure S3), and greater in the Northeast and Midwest regions compared with the others (Figure S4). Despite distinct seasonal patterns observed at many sites, imidacloprid was detected in stream water at least 50% of the time in all months among the 47 trend sites and ranged from 51% (March and April) to 81% (June) (Figure 4). The median monthly concentrations among the 47 trend sites exceeded the chronic benchmark in all months and ranged from 11.3 (February; mean = 17.3 ng/L) to 28.0 ng/L (July; mean = 60.0 ng/L), demonstrating a persistent potential threat to aquatic life. Imidacloprid was detected in at least 50% of the trend site samples in all years and ranged from 50% (2022) to 77% (2018), while the median concentration ranged from 12.9 (2013; mean = 25.0 ng/L) to 20.2 ng/L (2020; mean = 38.8 ng/L; Figure 4).

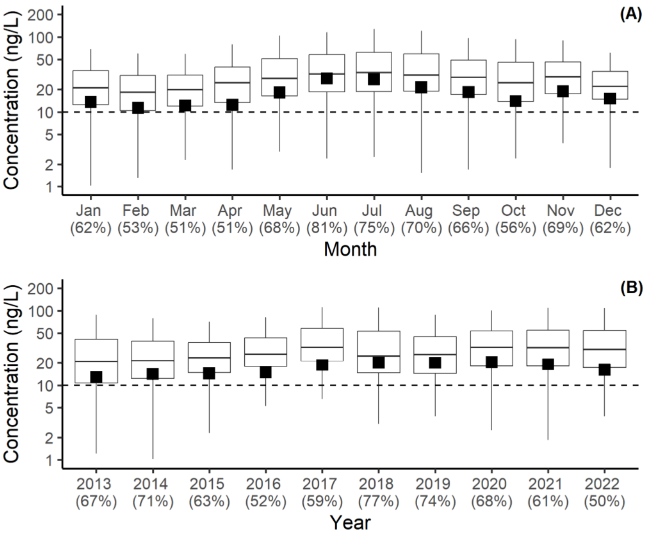

Figure 4. Detected imidacloprid concentrations arranged by (A) month and (B) year from 47 U.S. river sites eligible for trend analysis plotted with a log-scale *y*-axis. The box represents the interquartile range of detected sample concentrations with the median denoted with the middle line. The lower and upper lines extend 1.5 times the interquartile range from the top and bottom of the box. Percentages in the *x*-axis labels denote the overall detection percentage for each month and year. Large squares indicate the median monthly concentration adjusted for nondetect samples using the regression on order statistics method.[72] The horizontal dashed line denotes the U.S. Environmental Protection Agency chronic (10 ng/L) aquatic life benchmark for freshwater invertebrates.[37]

Previous studies have documented elevated imidacloprid concentrations during increased discharge periods,[30] which tend to coincide with spring months when crops are planted in many regions of the United States. Approximately half of the trend sites (n = 23) showed a positive relation between concentration and discharge as indicated by a concentration−discharge (C−Q) slope of >0.1, one-third (n = 16) showed little concentration variability with discharge (i.e., chemostatic; $|C-Q \text{ slope}| \leq 0.1$), and eight had a negative relation between concentration and discharge (C−Q slope < −0.1).[50] Large C−Q slopes occur when higher concentrations are observed during elevated discharge and suggest dominant sources that are transported through fast delivery pathways.[95] In contrast, chemostatic or negative C−Q slopes can indicate saturation with legacy stores that control the mobilization and transport of imidacloprid or the presence of point sources that lead to increased concentrations during periods of low streamflow. Improvements in sites with positive C−Q slopes can result from mitigation measures that target dominant delivery pathways and critical source areas.[95] However, short-term mitigation measures are less effective at reducing imidacloprid concentrations in watersheds with flat or negative C−Q slopes, which require long-term and large-scale mitigation approaches.[95] Major tributaries and mainstem study sites along the Mississippi River transitioned from positive to flat and negative C−Q slopes from upstream to downstream, which may indicate a downstream saturation of imidacloprid and an accumulation of legacy sources (Figure 5).

Rising Concentration Trends Signal Escalating Risks across All Geographic Contexts. Increasing trends in imidacloprid concentration and load were observed at most sites, land use types, drainage areas, and geographic regions and were of large magnitude, suggesting widespread contamination of nontarget ecosystems (Figures 1 and 6). Among the 47 sites that met criteria for trend analysis, most sites (74%; n = 35) reported a likely upward trend, three sites had a somewhat likely upward trend, three sites did not have an upward or downward trend, four sites reported a somewhat likely downward trend, and two sites had a likely downward trend from 2013 to 2022 (Figure 1).[50] Trends from uncorrected environmental samples were very similar to the trends reported from samples corrected for changes in laboratory reagent spike recovery,[50] with only seven out of 47 sites reporting minor changes in trend category. The trend, as a percentage change, was highly variable (mean +69.6%; standard deviation = 71.5%) and ranged from −68.6% (Zollner Creek near Mt Angel, Oregon [USGS site 14201300], hereafter Zollner Creek near Mt Angel) to +305.4% (Little Arkansas River near Sedgwick, Kansas [USGS site 07144100]). Fifteen sites reported a concentration percentage increase greater than 100%.[50] Based on concentration change, the mean trend increase exceeded the chronic benchmark for aquatic life (mean = 10.6 ng/L; standard deviation = 15.6 ng/L) but showed a wide range from −33.1 (Zollner Creek near Mt Angel) to +64.3 ng/L (Bogue Phalia near Leland). Eighteen sites reported a positive trend exceeding 10 ng/L.[50]

Most of the trend sites in each land use category, drainage area category, and geographic region had likely upward trends (Figure 6). Likely upward trends were reported for 84% of the agricultural sites, 67% of the developed sites, 71% of the mixed sites, and 33% of the undeveloped sites. Small watersheds had

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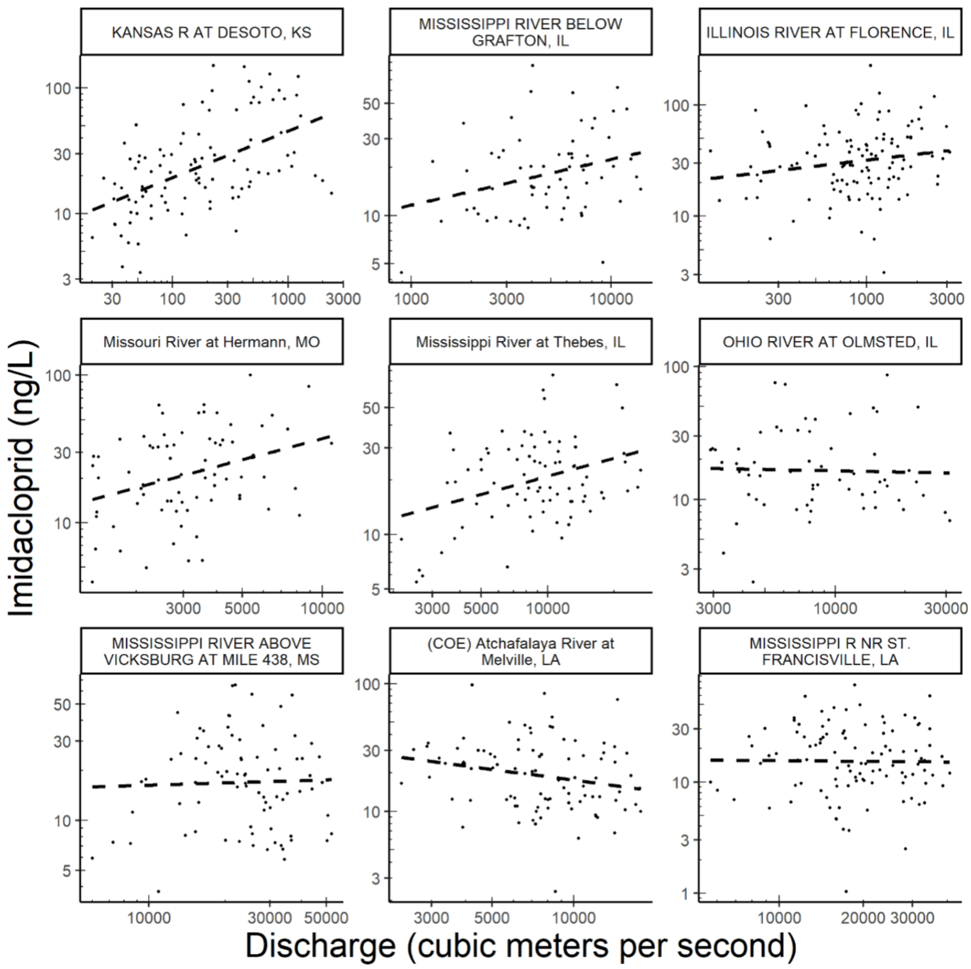

Figure 5. Imidacloprid concentration-discharge relations at the Mississippi River mainstem and tributary study sites in the United States from 2013 to 2022. The circles represent detected concentrations. The black dashed line is a linear model fit to the log-transformed concentration and discharge data. Plot labels denote the Location Name presented in the work by Miller et al.[50]

a likely upward trend at 65% of the sites, 73% of medium watersheds had a likely upward trend, and 92% of the large watersheds reported a likely upward trend including the outlet of the Mississippi River (Mississippi River near St. Francisville) which reported a +79.9% concentration increase (+9.0 ng/L). Regionally, 100% of Northeast sites, 88% of South sites, 78% of Midwest sites, 50% of Pacific sites, and 20% of West sites reported likely upward trends.

The median annual predicted load from the trend sites from 2013 to 2022 was 85.0 kg[50] and ranged from 0.17 kg (Little Cottonwood Creek at Jordan River near Salt Lake City [USGS site 10168000] – 2018) to 19,284 kg (Mississippi River above Vicksburg – 2019). Two sites representing the discharge of the Mississippi River into the Gulf of America (Mississippi River near St. Francisville and Atchafalaya River at Melville) both had likely upward trends, and the combined total (2013–2022) and mean annual imidacloprid loads were 129,489 kg (142.7 U.S. ton) and 12,949 kg/year (14.3 U.S. ton/year), respectively (Figure 7). The combined imidacloprid load from these two sites from 2013 to 2017 (53,060 kg), the most recent years with published agricultural use estimates,[63] represents 3.7% to 4.6% of the estimated agricultural use within the two watersheds, based on high and low estimates of 1,420,267 and 1,141,160 kg, respectively.[63] This mean annual load delivered to the Gulf of America may pose ecological risks to marine life and represents enough imidacloprid active ingredient to treat approximately 2,600 km^2 (1,000 mi^2) of corn assuming 2 grams of imidacloprid per kilogram of seed and 25 kilograms of seeds per hectare[96] or about 360 km^2 (139 mi^2) of turfgrass using an application rate of 360 grams per hectare.[97]

Large-Scale Patterns and Year-Round Presence Highlight Persistent Threats. Data collected from 2013 to 2022 from 77 sites located across the United States indicated year-round potential chronic exposure to imidacloprid for freshwater invertebrates, especially in watersheds draining agricultural and developed land. Sample concentrations exceeded the chronic benchmark in all months across all dominant land use, drainage area size, and geographic region categories. Results presented in this study suggest that many medium to large watersheds contain potentially toxic levels of imidacloprid throughout the year, especially in the South and Midwest. The year-round presence of imidacloprid may imply year-round applications or persistence in the environment that exceeds the expected degradation times in soil and water. A range of half-life times have been reported for imidacloprid in soil (40 to 1,669 days) and are influenced by pH, soil texture, fertilizer amendments, and organic matter content.[98,99] Increasing imidacloprid concentration trends may indicate accumulation from the increased use of the insecticide on agricultural and developed landscapes. From 2013 to 2017, the high-estimated annual agricultural imidacloprid use declined in the United States from 973,489 to 637,841 kg;[63] however, beginning in

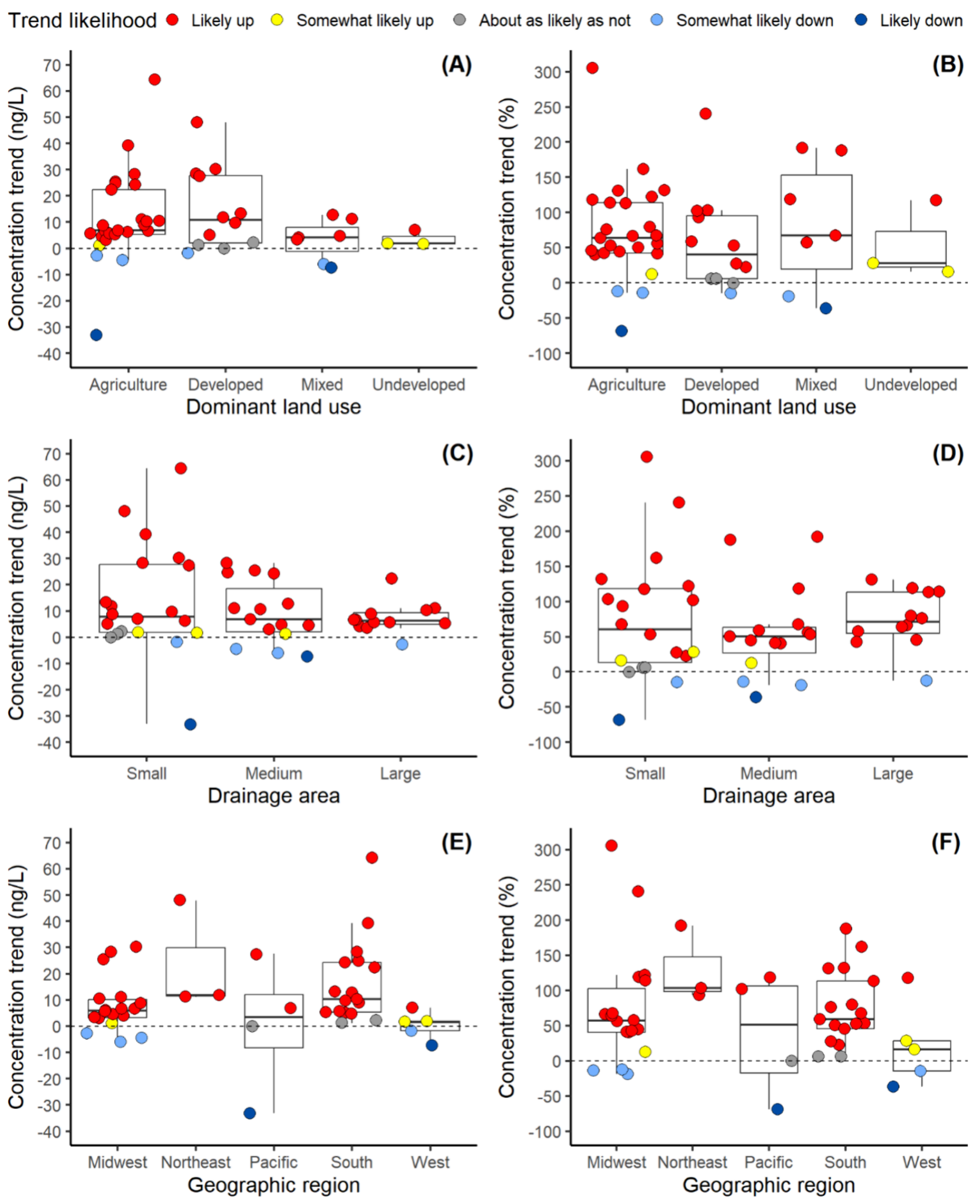

Figure 6. Imidacloprid concentration trend results for 47 U.S. river sites arranged by (A, B) dominant land use, (C, D) drainage area, and (E, F) geographic region. Trend results are presented in concentration units (A, C, E) and percentage change (B, D, F) from 2013 to 2022 from the trend study sites and colored by trend likelihood.

2015, the data provider discontinued making estimates for seed treatment application of pesticides,[63] which likely explains the large decline between 2014 (1,054,158 kg) and 2015 (467,987 kg). In addition, the lack of pesticide use data for non-agricultural purposes makes it challenging to compare pesticide use patterns with changes in concentrations detected in the environment.

Frequent imidacloprid detections outside the growing season in medium to large sized watersheds, notably the Mississippi River which became more chemostatic in the downstream direction, may suggest that the insecticide is being transported long distances beyond the targeted use locations, resulting in potential saturation of groundwater systems by imidacloprid at large scales.[31] Imidacloprid is highly soluble and moderately mobile and, thus, has the potential to leach into groundwater and be a legacy source of surface water contamination[99] from groundwater discharge, creating a persistent threat to ecosystem health. While this study solely focused on imidacloprid, samples collected between 2013 and 2017 from this network indicate mixtures of insecticides, herbicides, and fungicides are commonly observed in most samples, with 88% of samples indicating five or more pesticides,[39] potentially increasing the exposure risk from additive or synergetic effects.

The results of this study showed that imidacloprid contamination impacts freshwater and estuarine water resources, and they are aligned with results from other recent global studies.[100−104] In France, imidacloprid was the main neonicotinoid used between 2005 and 2018. Water contamination risk, measured as the percentage of sites with at least one sample exceeding 20 ng/L, increased during 2005−2018 (2018 peak at 22% of sites), decreased sharply following the 2018 ban for all outdoor crops in the European Union with exceptions for emergency use, but it remained detectable in rivers after four years.[103] 28% of the samples in the present study exceeded 20 ng/L, and 84% of the study sites had at least

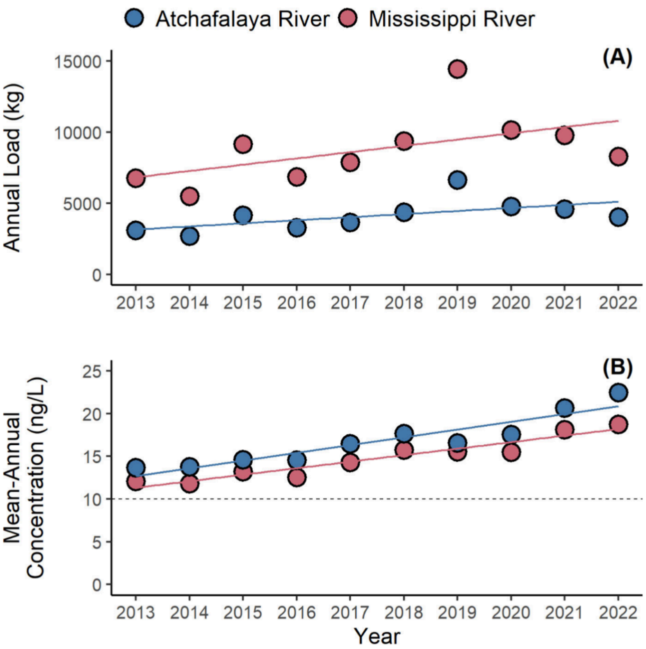

Figure 7. Predicted (A) annual load and (B) mean annual concentration of imidacloprid for the Mississippi River near St. Francisville and Atchafalaya River at Melville sites representing the outlet of the Mississippi River into the Gulf of America. The dashed horizontal line denotes the U.S. Environmental Protection Agency chronic (10 ng/L) aquatic life benchmark for freshwater invertebrates.[37]

one sample exceeding 20 ng/L. No current national restrictions for imidacloprid exist in the United States. Imidacloprid was detected in 54% of samples in rivers draining to the Great Barrier Reef of Australia[104] (median concentration = 11 ng/L), and significant increases in concentration were determined from six out of 14 sites affected by agricultural and developed sources, similar to the results presented here. In China, which has among the highest levels of pesticide usage globally, imidacloprid is frequently detected in agricultural soils, the atmosphere, streams draining both agricultural and urban areas, as well as adjacent estuaries and oceans, prompting extensive research on the human and ecological risks of imidacloprid and neonicotinoid exposure.[101,102] The mean annual imidacloprid load to the Gulf of America from the Mississippi River estimated in this study (12.95 t) was more than twice the reported load to the South China Sea (5.80 t) and more than a third of reported load to the East China Sea (34.61 t).[101]

This national, long-term assessment of streams and rivers in the United States revealed widespread imidacloprid contamination. The year-round presence of imidacloprid above ecologically relevant thresholds occurred consistently across the nation for a decade. This persistence is likely causing chronic impairment to sensitive freshwater invertebrates and could present challenges to water managers attempting to improve water quality conditions in impaired streams and rivers. The widespread use of imidacloprid for a variety of agricultural and nonagricultural purposes makes it difficult to pinpoint a specific source of increasing trends. In fact, the data in this national assessment suggest both agricultural and nonagricultural sources have contributed to the widespread detections and increasing concentrations of imidacloprid in rivers across the United States. Increasing imidacloprid concentrations and loads, particularly from large watersheds, demonstrate transport far beyond intended target locations and pose long-term concerns for human and ecosystem health. The findings of this study underscore the importance of continued monitoring and research that captures the full complexity of neonicotinoid use and transformation, including the presence of multiple compounds and their transformation products,[105] to better understand environmental patterns and their potential implications for ecosystem health at national scales.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.5c07311.

Laboratory quality assurance samples and additional imidacloprid results from environmental samples (PDF)

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. S.A.M. conducted the formal analysis and made the visualizations. S.A.M. and T.S.S. wrote the original draft. All authors participated in relevant scientific discussions and provided comments on the manuscript.

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Notes

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