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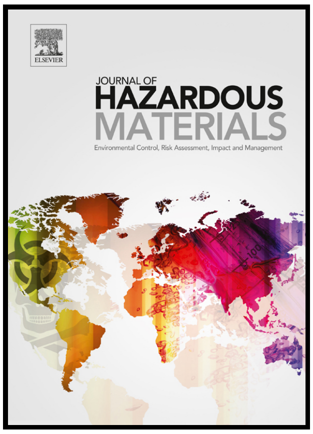

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Beyond Agriculture: Land Use Thresholds Governing Pesticide Mixture Risks in Megacity Surface Waters

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Abstract

Growing concerns have emerged regarding the risks of pesticide mixtures in surface water ecosystems, yet the mechanisms through which human activities, especially land use patterns, affect these risks remain inadequately studied. This research presents an innovative approach, combining multi-scale land use analysis with pesticide risk assessment, quantifying relationships between mixed pesticide ecological risks and land use patterns. Findings indicate that the impacts of urban land use on pesticide ecological risks surpass the traditionally recognized agricultural effects, demonstrating significant spatial scale-dependent effects. Generalized additive model analysis reveals that 1−3 km and 2−3 km buffer zones represent the critical ranges where urban land use and cropland, respectively, have significant impacts on pesticide risks. Non-parametric change point analysis determined critical land use thresholds triggering significant ecological risk increases: 10−25% for cropland and 10−30% for urban areas. These discoveries provide crucial quantitative foundations for landscape

planning and pesticide risk management. The results not only challenge traditional views of agricultural activities as primary pesticide sources but also provide new perspectives for pesticide pollution control and water quality management in large cities.

Keywords
Pesticide pollution, mixed ecological risk, land use, ecological thresholds, landscape management

1. Introduction

As a key indicator of the Anthropocene [1], pesticide pollution has emerged as a global environmental challenge. Most parent pesticides worldwide enter environmental media such as water and soil through spray drift, leaching, plant uptake, and runoff, leading to extensive environmental contamination [2]. Globally, 74.8% of agricultural land faces potential pesticide pollution risks, with a concerning 31.4% classified as high-risk [3-5]. Previous research has focused on analyzing sustainable pesticide use strategies [6-8], evaluating toxicity-based priority control lists [9, 10], and analyzing the spatiotemporal distribution and fate of pesticides [11, 12]. With advancing research, the "source tracing" of pesticides has increasingly emerged as a key research priority [13-15]. Investigating factors affecting pesticide risks, whether human activity elements or natural factors, has gradually become an important part of "source tracing" research and potentially yields substantial ecological benefits [15]. Nevertheless, there is limited research analyzing the effects of human activities on pesticide pollution, particularly regarding how land use planning influences pesticide transport into water bodies and the subsequent ecological risks to aquatic organisms. Consequently, there is a pressing need to quantify the influence of human activities on pesticide risks, enabling source control of pesticide threats to ecosystems.

Investigation of factors affecting pesticide risks first requires assessment of the ecological risks caused by pesticides. Assessment of potential pesticide risks to ecosystem organisms typically provides guidance for pesticide control measures [16]. However, pesticide detection, registration, use, and traditional ecological risk assessments primarily focus on single active compounds [17], relying on single-species laboratory exposure and toxicity data [18], while real environmental exposure occurs in mixtures, often presenting greater actual risks to aquatic ecosystems than estimated [19, 20]. It is therefore imperative to assess the ecological risks of mixed pesticide exposure to gain a deeper understanding of their environmental consequences.

The quantitative assessment of mixed pesticide risks directly reflects the influence of various factors on pesticide risks [21, 22]. According to existing research, human activities, especially land use intensity, represent critical factors influencing pesticide exposure and ecological risks [23]. Changes in land use and land cover, which significantly alter Earth's energy balance and biogeochemical cycles, are major drivers affecting ecosystem health [24, 25]. Research [26] has shown that when forests and pastures are converted to farmland, soil organic carbon decreases by 24% to 59%, leading to soil nutrient loss and ecological function degradation. Gossner et al. [27] observed species from 12 trophic groups and found that increased land-use intensity significantly reduced the biodiversity of many different trophic groups. Pesticide exposure levels in intensive agricultural zones and densely populated areas substantially exceed those in natural landscapes such as forests and aquatic ecosystems [15], demonstrating that both agricultural activities and urbanization contribute to pesticide contamination. However, current research primarily focuses on agricultural activities' effects on pesticide presence and distribution in surface waters [3, 28-30], often neglecting urban pesticide pollution [5], from sources like urban landscaping, rural tourism, and fruit/vegetable cultivation areas [31]. Furthermore, active pesticide ingredients typically show higher concentrations in non-agricultural environments [32], with multiple studies reporting higher insecticide levels in U.S. urban surface waters than in agricultural areas [31, 33, 34]. Different land use types significantly affect pesticide migration and transformation in the environment [35], while rational watershed-level land use distribution through landscape planning offers a novel approach to pesticide pollution analysis [36]. Pesticide risk minimization can be achieved through enhanced protection of ecologically vulnerable areas and pesticide-sensitive buffer zones, reduced land degradation risk, and improved land use efficiency [37, 38]. Rational land use planning can thus yield greater ecological benefits [39]. Currently, there is a global shortage of methods for pesticide source control through landscape-scale land use pattern planning. Studies examining how pesticide ecological risks respond to land use patterns remain lacking [36, 40].

Based on these scientific issues, this study proposes a novel approach combining multi-scale land use analysis with pesticide risk assessment. Response relationships between land use patterns and mixed pesticide ecological risks are established across different buffer zones, employing generalized additive models (GAM) and non-parametric change point analysis (nCPA) to quantitatively identify key scales and thresholds. Beijing, covering 16,410 square kilometers with 21.5 million inhabitants, ranks as the world's eighth-largest megacity [23]. Beijing exhibits substantial spatial heterogeneity in population density, varying from hundreds to over 20,000 people per square kilometer from suburbs to downtown, with significant variations in human disturbances, activity frequency, and complex land use patterns [23, 41]. These complex land use patterns and marked spatial heterogeneity provide an ideal platform for examining landscape effects on pesticide pollution.

The study aims to: (1) Quantify ecological risk levels and spatial distribution patterns of pesticide mixtures in megacity surface waters; (2) Identify and validate mechanisms by which urban and agricultural land use influence pesticide ecological risks; (3) Establish the critical buffer zone ranges and threshold levels of land use that influence pesticide risks. This research will provide a scientific basis for challenging traditional agriculture-centric views of pesticide pollution while introducing new approaches to landscape planning-based pesticide risk management. Other cities may optimize the distribution and proportions of impervious surfaces, cropland, forests, and grasslands based on their development objectives, or modify buffer zone extents according to current land use type proportions and pesticide risk status, to implement source-based pesticide risk control. Research innovations include: the first exploration of spatial response mechanisms between land use and mixed pesticide risks at megacity scale; introduction of buffer zone-based risk assessment methodology; and quantification of critical land use thresholds triggering significant ecological risk increases.

2. Material and method

2.1. Study area and collection of samples

Beijing exhibits substantial spatial heterogeneity in population density, with significant variations in human disturbances, including land use patterns. Between 1992 and 2008, more than half of agricultural land was converted to urban and industrial use [41], with urbanization reaching 86.6% by 2019 [42]. Intensive human activities have resulted in significant pesticide contamination of surface waters [43]. Additionally, Beijing has complex water systems including rivers and large reservoirs, with water quality issues becoming increasingly prominent due to extensive groundwater extraction, rapid population growth, and fast socioeconomic development [44, 45]. Beijing thus presents an ideal area for investigating pesticide sources, distribution patterns, and risk variations [23].

To investigate pesticide concentrations, spatiotemporal distribution characteristics, ecological risks, and their driving factors in the surface waters of rivers, lakes, and reservoirs across Beijing, two surface water sampling events were carried out. Sampling occurred in November 2020 (dry season) and April 2021 (normal season), for the following reasons: (i) it coincided with the end of one crop growing season and the beginning of another, making it ideal for assessing the extent of pesticide effects on organisms; (ii) no insecticides were used in the short term, which facilitated the identification of insecticide types that are frequently found in the environment, as they exhibit accumulation and persistence characteristics. As illustrated in Figure 1(b), a total of 60 sampling points were established, and specific details on sampling methods and sample pretreatment can be found in Text S1.2.

2.2. Chemical analysis

Reference standards and internal standards for 49 target compounds across 4 categories were obtained from Alta Scientific Co. Ltd (Tianjin, China) (refer to Text S1.1 and Table S2 in the supplementary materials). Analysis was performed using Waters ACQUITY liquid chromatograph and Xevo T-QS triple quadrupole mass spectrometer (ESI-MS/MS, Waters Co., Milford, MA, USA), following prior studies conducted by the research group [23, 46, 47]. Emerging pollutants were ranked by their ecological risks to aquatic organisms, with neonicotinoid (NEOs) and organophosphate pesticides (OPPs) ranking among the highest. This study therefore focused on measuring concentrations of 9 NEOs, 14 OPPs, 15 triazine pesticides (TPs), and 11 carbamate pesticides (CAs). Chromatography-grade organic solvents were obtained from Thermo Fisher Scientific (USA). Chromatographic grade formic acid was purchased from Sigma-Aldrich (St. Louis, Missouri). Milli-Q water was generated by a Milli-Q system (Millipore, Bedford, USA). Analyzed compounds are listed in Table S1, while analytical methods and instrumental details are provided in Table S2. Details of quality assurance and quality control measures can be found in Text S1.4 and Table S3.

2.3. Ecological risk assessment

Experimental values and predictions were obtained from the ECOSAR model [46, 48]. The principles for screening toxicity data are provided in Text S3. When several experimental values are available, the minimum value is chosen to ensure that all potential risk substances are included [49]. Toxicity Units (TU) were employed to assess the potential ecological risks posed by each pesticide and mixed pesticides to aquatic organisms in urban surface waters [47], with the formula as follows:

$$TU = {}^{C}/_{PNECs} \tag{1}$$

In this context, the predicted no effect concentration (PNEC) is defined as the minimum toxicity threshold for the most sensitive group of species, indicated by chronic risk thresholds (CRT). *C* represents the concentration of the pollutant, including the average concentration. The formula for calculating the risk of mixed pesticides [47] at each sampling point is as follows：

$$TU_{S_x} = \frac{\sum_{i=1}^{n} TU_{CRT_i}}{i} \tag{2}$$

Where TU_{Sx} denotes the chronic combined risk of compound *i* at the sampling point. Prior research categorized TU results into three levels: $TU_{Sx} < 0.1$ indicates that the risk is negligible, $0.1 < TU_{Sx} \leq 1$ indicates low risk, $1 < TU_{Sx} \leq 10$ indicates a moderate risk, and $TU_{Sx} > 10$ indicates high risk.

2.4. Land use data acquisition and processing

Land use data were extracted from the 2019 China land cover dataset (CLCD) with a spatial resolution of 30 meters, provided by Wuhan University [50]. As illustrated in Figure 1b. Land use types include arable land (e.g., rice paddies and dry fields), forests, grasslands, water bodies (e.g., rivers, ponds, and reservoirs), impervious surfaces (e.g., residential, industrial, and mining areas), and unused land (e.g., deserts, wetlands, and bare land). Circular buffer zones of 0.1, 0.2, 0.5, 1, 1.5, 2, 2.5, 3, 5, and 10 kilometers were established around each sampling point using ArcGIS 10.8, to extract land use type data for these buffers (Table S9), along with the specific area proportions of different land use types, which reflect the spatial layout of land types within the various buffer scales.

2.5. Statistical analysis

One-way ANOVA was employed to assess significant differences among variables. The Kolmogorov-Smirnov D statistical test was applied to check the distribution of pesticide residual concentrations in water. T-tests were utilized to compare two independent samples. For comparisons involving more than two independent samples, one-way ANOVA was applied. Pearson's correlation test was used to assess the linear relationship between two variables. All the statistical analyses were performed using R (version 4.2.3). The significance level for statistical tests (p-value) was established at 0.05. Spearman's correlation coefficient was used to compute the linear correlation between the mixed ecological risks of pesticides and the proportions of land use types. First, Box-Cox transformations were applied to the areas of arable land and impervious surfaces, along with the ecological risk data, to analyze the interactions between the two influencing factors. Positive Matrix Factorization (PMF) was applied to identify and characterize the principal sources of pesticide contributions. GAM was used to establish the relationship between explanatory variables and response variables across different buffer scales. GAM is a non-linear additive regression model for the response variable Y [51], expressed as follows [52, 53]:

$$g\big(E(Y_i)\big) = \alpha + s_1\big(X_{1,i}\big) + s_2\big(X_{2,i}\big) + \cdots + s_p\big(X_{p,i}\big)$$

(3)

Where Y_i denotes the mixed ecological risk TU_{Sx} at the *i-th* sampling point, X_i is the explanatory variable for the *i-th* sampling point, p indicates the total number of explanatory variables (corresponding to the number of buffer scales), α represents the model intercept, s(·) is the smoothing function applied to the explanatory variable X (capturing non-linear relationships). $g(\cdot)$ is a link function; in this study, an identity link function was utilized. The model evaluation was conducted using sample size (n), explained variance (R^2), and minimized generalized cross-validation scores. The "*mgcv*" and "*ggplot2*" in R (version 4.2.3) were utilized, and the results are shown in Table S10.

2.6. Ecological threshold derivation

Ecological thresholds were derived by identifying change points in environmental parameters and landscape metrics using nonparametric change-point analysis (nCPA) [54-56]. Initially, observations from multiple sampling points were ordered along a gradient, a threshold or sudden change in the statistical attributes of the dependent variable occurs in the relationship between the explanatory variables and response variables, enabling the identification of change points along the landscape metric gradient that lead to shifts in ecological risk [57]. Let $y_1, y_2, \ldots, y_n$, denote the sequence of ecological risk variables observed along the landscape metric gradient, with the nCPA calculation method given as follows:

$$D = \sum_{k=1}^{m} (y_k - \mu)^2$$

(4)

Where D represents the deviation, m is the sample size, and μ is the average of the m observations y_k.

Let i be the interval point between the two groups. The deviation redundancy Δi is calculated using Equation (5), and Δi for each possible change point t ($1 \le t \le n$) is always greater than or equal to 0. The value of i that reaches the maximum Δi is identified as the change point t.

$$\Delta i = D - (D_{\le i} + D_{>i})$$

(5)

Where D、$D_{\leq i}$ and $D_{>i}$ represent the deviations for the data sets $y_1, y_2, \ldots, y_n$、$y_1, y_2 \ldots y_i$ and $y_{i+1} \ldots y_n$, respectively.

In general, actual observational data are limited; thus, nCPA is combined with the bootstrap method to estimate the frequency distribution of change points [58]. As a result, this research emphasizes the examination of changes in arable land and impervious surfaces. We used the bootstrap method to extract random samples of 1,000 pesticide mixture risk parameters and key landscape indicator datasets to compute the probabilities of change point occurrences using nCPA. All analyses were conducted in R (version 4.2.3) using the "*dplyr*", "*changepoint*" *an*d "*boot*" packages.

3. Result

3.1. Seasonal and spatial changes in pesticide concentrations and ecological risks

A total of 48 pesticide compounds were identified in Beijing's surface waters. During dry and normal seasons, total pesticide concentrations ranged from 73.17−3412.95 ng/L and 45.17−4211.79 ng/L, with respective mean values of 566.76 and 704.77 ng/L. Dinotefuran exhibited notably higher concentrations compared to other pesticides, ranging from 0.04−2169.37 ng/L and 0−2628.38 ng/L in dry and normal seasons, respectively (with mean values of 272.75 and 349.97 ng/L). Seasonal variations in pesticide concentrations were evaluated using independent sample *t*-tests, while spatial distribution patterns were analyzed using Kruskal-Wallis one-way analysis of variance. The results (Table S5) demonstrate significant spatial variations ($p < 0.05$) in total concentrations and levels of NEOs, TPs, and CAs (Figure 1c and 1d). Conversely, OPPs exhibited significant seasonal variations but showed no significant spatial distribution differences. Mean pesticide concentrations in eastern and southern Beijing (Tongzhou, Chaoyang, and Changping districts) during dry and normal seasons (2113.90 and 2451.74 ng/L, respectively) were fourteen times higher than those in western and northern regions (Miyun and Huairou districts: 146.45 and 175.57 ng/L, respectively). The primary distinction between these regions lies in the intensity of human activities, with results indicating that both during dry and normal seasons, densely populated areas exhibited higher pesticide concentrations and detection frequencies, whereas regions with extensive forest coverage or mountainous areas typically showed lower detection levels. Pesticide composition distributions across different sampling locations in Beijing (including southeastern Tongzhou, Daxing, and Chaoyang districts, and western/northern Yanqing, Huairou, and Miyun districts) were matched with regional crop patterns to determine source attribution, with PMF analysis revealing distinct compositional differences among source factors (Table S8). The model results indicate that grain crops, fruit and vegetable crops, and other crops constitute the primary contributing sources of pesticides. Detection variations across different land use types directly demonstrate the influence of spatially heterogeneous human activities on pesticide contamination.

As illustrated in Figure 2, mixed risk TU_{Sx} values ranged from 0.17 to 14.37 during the dry season, with 32% of sampling sites showing elevated risk levels, and from 0.13 to 24.04 during the normal season, with 35% of sampling sites showing elevated risk levels. Significant spatial variations in TU_{Sx} were observed across sampling locations ($p < 0.05$), with high-risk sites predominantly concentrated in urban clusters and southeastern regions characterized by intensive agricultural activities. These findings clearly demonstrate the contributions of agricultural activities and urbanization to elevated ecological risk levels. To assess risks associated with different pesticide types, TU_{Sx} values were calculated for four pesticide categories, as shown in Figure S2. NEOs exhibited the highest ecological risk, with TU_{Sx} values ranging from 0 to 78.85 and 0 to 136.86 in the dry and normal seasons, respectively, followed by OPPs with TU_{Sx} values of 0.55 to 2.47 and 0.24 to 3.40 in dry and normal seasons. These findings demonstrate that NEOs and OPPs are the primary contributors to mixed ecological risks from pesticides in surface waters.

3.2. Relationship between land use patterns and ecological risks

Previous research has established that pesticide ecological risks correlate with natural climatic conditions, human land use patterns, pesticide physicochemical properties, and emission factors, with human land use exerting the predominant positive influence on ecological risks [23]. We conducted further analysis to examine the relationships between various land use types and pesticide ecological risks. The land use types were classified into Cropland, Forest, Shrub, Grassland, Water, Barren, and

Impervious surfaces, and we analyzed Spearman correlation coefficients between mixed ecological risks and individual pesticide ecological risks with different land use types across multiple buffer zones (0.1, 0.2, 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 5.0, and 10.0 km). The results (Figure S4 and Table S7) indicate significant positive correlations between TU_{Sx} and both cropland and impervious surfaces, with correlation coefficients ranging from 0.04 to 0.61 and 0.20 to 0.72, respectively, across different buffer zones. This directly demonstrates the substantial contribution of both cropland and urban area proportions to ecological risk. Conversely, forests and grasslands exhibited significant negative correlations with TU_{Sx}, with correlation coefficients ranging from −0.79 to −0.44 and −0.78 to −0.23, respectively, demonstrating their significant mitigating effect on pesticide-related ecological risks to aquatic organisms.

Furthermore, the results reveal that across all buffer zones, correlation coefficients between impervious surfaces and TU_{Sx} consistently exceeded those between cropland and TU_{Sx} at corresponding scales. To elucidate the relationship between these two influential factors and identify the primary driver of mixed pesticide risks, we conducted interaction and individual effect analyses on TU_{Sx}, cropland, and impervious surface data, as presented in Table 1. Cropland and impervious surfaces demonstrated synergistic effects with highly significant interactions ($p < 0.001$), with the interaction term exhibiting a positive influence on TU_{Sx}. The synergistic effect of cropland and impervious surfaces reduces the proportion of forests and grasslands, potentially intensifying surface runoff and facilitating the convergence of complex pesticide mixtures in surface water, thereby exacerbating mixed pesticide contamination [59]. The individual effect analysis revealed that impervious surfaces exerted a more significant influence on TU_{Sx} than cropland ($p < 0.001$). Thus, synthesizing the results from both interaction and individual effect analyses, while cropland and impervious surfaces demonstrate synergistic effects, impervious surfaces exert a stronger influence on mixed risks.

Table 1 Individual effects of arable land and impervious surfaces on ecological risk along with interaction analysis

	Estimate	Std. Error	*t* value	*P*(>|t|)
(Intercept)	−2.472	0.232	−10.642	<0.001
Cropland	0.190	0.051	2.156	<0.050
Impervious	0.420	0.053	7.901	<0.001
Cropland: Impervious (synergistic)	0.098	0.023	——	<0.001

3.3. Scale-dependent effects of land use on ecological risks

The impact of cropland and impervious surfaces on mixed risks varies depending on buffer zone extent. Spearman correlation coefficients for both factors showed minimal variation at buffer zones of 0.1−0.5 km (Table S7); thus, we focused on buffer zones of 1−5 km to more precisely evaluate the scale-dependent effects of land use on ecological risks. GAM was fitted using mixed risks (TU_{Sx}) during dry and normal seasons at various sampling points as response variables, with cropland and impervious surface proportions at different buffer scales serving as explanatory variables. We analyzed the nonlinear influence of land use on TU_{Sx} at optimal scales. Fitting results shown in Figure 3a and 3c indicate that when cropland served as the explanatory variable, mixed ecological risks exhibited minimal variation with increasing cropland proportion at buffer zones of 1.0 km and 1.5 km.

Within 2−3 km buffer zones, nonlinear relationships emerged, with significant increases in mixed ecological risks occurring after cropland proportion exceeded 40% in 2 km zones, while at 2.5 km zones, dry and normal seasons demonstrated decreasing and slightly increasing trends, respectively. TU_{Sx} showed sudden increases or decreases when cropland proportion exceeded 60%, indicating instability in mixed risks at this buffer zone range. Beyond the 5 km buffer zone, increased impervious surface proportion resulted in stable or decreased TU_{Sx} trends. Figures 3b and 3d illustrate the response of TU_{Sx} to impervious surface proportion, demonstrating nonlinear relationships across all buffer zone scales. TU_{Sx} exhibited significant fluctuations across broader ranges (1−3 km); within 1 km buffer zones, it showed gradual decreases with increasing impervious surface proportion; at 3 km buffer zones, dry and normal periods demonstrated increasing and increasing-then-decreasing trends respectively; at 5 km buffer zones, TU_{Sx} either maintained stable or decreasing trends with increasing impervious surface proportion. Significant changes in fitting trends between TU_{Sx} and both cropland and impervious surface area occurred within 2−3 km and 1−3 km buffer zones, respectively, while TU_{Sx} showed minimal variation and gradual increases within 1 km and approaching 5 km buffer zones. Thus, the 2−3 km and 1−3 km ranges represent the critical scale ranges for the impacts of cropland and impervious surface area on mixed risks, respectively, with differing trends between dry and normal seasons suggesting seasonal influences on scale effects. Land use planning strategies must account for critical buffer zones vulnerable to human land use impacts, enabling the achievement of desired ecological benefits cost-effectively.

3.4. Analysis of change points for key land use types impacting mixed ecological risk

The goal of human land use planning is to reduce mixed pesticide risks. Consequently, this study examined threshold values where relationships between mixed risks and human land use proportions undergo sudden changes. Drawing from GAM results and Spearman correlation coefficients (Table S7 and S10), we utilized cropland and impervious surface areas at the 2 km buffer zone scale to determine ecological risk transition thresholds. Figure 4 illustrates the distribution of mixed ecological risk change points along gradients of cropland and impervious surfaces, derived from 1000 bootstrap simulations using the nCPA method. From a risk assessment standpoint, cumulative distribution plots provide direct probability estimates of sudden risk changes. Key interval values for mixed risk transitions correspond to cropland area proportions of 22−25% (Figure 4a and 4c). With cropland proportions below 22%, cumulative mixed risk remains under 60% and increases gradually; when cropland proportion exceeds 30%, the cumulative probability of sudden mixed risk increase surpasses 90%. The critical threshold range for impervious surface proportion triggering mixed pesticide risk transitions is 25−30% (Figure 4b and 4d); beyond approximately 35% impervious surface coverage, the cumulative probability of sudden mixed risk increase reaches 100% across all seasons. Furthermore, mixed risk remains at low or negligible levels when cropland or impervious surface proportions are below 10%.

4. Discussion

4.1. Temporal and spatial dynamics of pesticide contamination in metropolitan river systems

Pesticides are prevalent in cities worldwide [60]. A review of 228 pesticides monitored in global urban rainwater revealed 85 frequently detected compounds, with diuron, simazine, atrazine, metolachlor, and 2,4-D as the top five. These pesticides showed median concentrations below 100 ng/L but high detection frequencies [4]. Analysis of rainwater samples across 17 U.S. states revealed concentrations of widely used agricultural herbicide atrazine and insecticides imidacloprid and fipronil typically ranging from 3 to 300 ng/L [60]. The spatial variation in pesticide contamination correlates strongly with the intensity of regional human activities [61], potentially explaining spatial differences in pesticide ecological risks.

Significant spatial variations were observed in TU_{Sx} and mixed ecological risks of NEOs, TPs and CAs, though seasonal variations were less pronounced. The overall mixed ecological risk exhibits a gradient pattern, increasing from west and north to southeast, with high-risk areas concentrated in the southeastern region, likely attributable to variations in land use patterns. Land use type variations affect the differential risks that pesticides pose to aquatic organisms. The southeastern region of Beijing features concentrated residential and commercial areas bordered by cropland [62], characterized by intense human activity [63], resulting in high pesticide inputs that significantly impact the health of river and lake ecosystems. These findings are consistent with the research of Ana et al. [64], which indicates that the spatial distribution of pesticide is primarily influenced by regional human activities, including agricultural production and urban usage, where land use changes driven by intensive human activity increase pollutant inputs, potentially degrading water quality and elevating ecological risks [65, 66]. The western and northern regions of Beijing, characterized by high forest coverage [67] and minimal human interference, experience lower pesticide inputs, while forest vegetation's ability to absorb and purify pollutants further reduces pesticide outputs in these areas [68, 69].

Crop types also indirectly influence ecological risks, with pesticide types, application methods, quantities, and frequencies varying by crop type. Differences in pesticide utilization rates among crops lead to varying ecological risks and spatial distribution patterns. Beijing's local statistical yearbook (Table S8) shows crop planting area proportions in descending order: grain crops > fruits and vegetables > other crops. PMF model was applied to analyze sources of two frequently detected pesticides (NEOs and OPPs) at high concentrations, as illustrated in Figure S3. Contribution rates of each pesticide type were matched with crop planting areas to generate Table S8, and analysis of major crops in different regions revealed primary sources of ecological risks across areas. In southeastern Beijing, where vegetable cultivation predominates, commonly used pesticides include NEOs (chlorantraniliprole, thiacloprid, and clothianidin) [11, 70] and OPPs (fenthion, methamidophos, and dimethoate) [71, 72], contributing to elevated ecological risks.

Seasonal and spatial variations in pesticide concentrations and ecological risks may also be attributed to physicochemical properties and application patterns. Most OPPs, being highly water-soluble, demonstrate strong migration and transformation capabilities in environmental waters through "discharge-redistribution-diffusion" processes [73], resulting in minimal spatial distribution variations. Studies on pesticide poisoning epidemiology [74] reveal distinct seasonal patterns in OPPs use, with increased usage correlating with higher summer and autumn pest pressures [74], partially explaining elevated mean ecological risks during autumn and winter (dry season).This results in varying ecological risks among pesticides. CAs show higher mixed risks in spring (normal season) than in the dry season, likely due to application timing, as research indicates seed-period pesticide applications result in significantly higher surface water concentrations during spring and autumn compared to summer and winter [75].

4.2. Key land use types driving pesticide risk

The influence of impervious surfaces on pesticide ecological risks surpasses that of agricultural land, challenging conventional wisdom. Traditional perspectives consider human activities, particularly large-scale intensive agriculture, as primary pesticide contributors [76]. This study reveals that agricultural land's contribution to surface water pesticides is less significant than expected, similar to findings from neonicotinoid studies in the Yangtze River Basin showing higher pesticide concentrations in urban versus rural areas [70]. Similar conclusions were reached by Gilliom [77] and Stone [78], indicating that pesticide persistence and surface water entry potential largely depend on pesticide type, usage quantity, timing, and soil characteristics [35, 79]. Agricultural pesticide usage follows fixed patterns with marked seasonal variations, peaking during spring and summer growing seasons compared to autumn and winter [80]. Large soil areas retain pesticides, enabling long-term diffusion and biochemical interactions with soil organic matter [81, 82], promoting pesticide degradation and reducing water contamination. Neonicotinoid compounds, for instance, with long soil half-lives (50−545 days) and K_{oc} of 56−225 L/kg, demonstrate high leaching potential [83].

The emergence of cities as zones of pesticide exposure and ecological risk potentially constitutes a global concern [84]. The expansion of urban and residential development leads to increased pesticide use in both quantity and variety within public facilities [30]. Substantial quantities of diverse pesticides are used in park landscaping, roadside vegetation management, residential greening, and non-agricultural applications such as disinfection, pest control, and construction materials [85]. Research suggests urban pesticide levels may correlate with economic development [86]. The rapid expansion of urban landscaping, a crucial component of urban development, typically results in increased government investment and consequently higher pesticide usage. Compounding the problem, many urban pesticide applicators lack professional training. As a result, insecticides are often overused for weed control [87]. Unlike seasonal agricultural applications, year-round urban landscaping maintenance leads to excessive pesticide use to maintain long-term pest control efficacy.

Urban pesticide contributions to rivers are linked to urban surfaces and drainage systems. High proportions of impervious surfaces lead to rapid surface runoff formation with simplified hydrological pathways and minimal transmission losses [88], directing pesticides into water bodies or initial rainfall retention facilities [89]. However, many densely populated and rural areas lack proper initial rainfall collection infrastructure, ultimately leading to discharge into surface waters. A study [4] reviewing 116 publications revealed pesticides as the predominant organic compounds in urban rainwater, constituting 36.3% of all detected organic substances. Furthermore, even when surface runoff reaches treatment facilities, technical limitations prevent urban wastewater treatment plants from reducing pesticide concentrations below risk thresholds prior to discharge [90, 91], potentially making these facilities point sources of pesticide pollution. Urban wastewater treatment plants may even serve as point sources of pesticide pollution.

Additionally, the distribution of pesticides in urban areas is influenced by their physicochemical properties; for instance, NEO pesticides easily transfer to surface water via runoff or rainfall erosion due to their low volatility and high water solubility [92]. Urban surface runoff, typically rich in organic matter [61], facilitates the adsorption and transport of hydrophobic and lipophilic pesticides [93], leading to a broader spectrum of pesticides originating from urban areas.

4.3. Scale-dependent effects and optimal buffer zones for risk management

The distance of buffer zones directly impacts pesticide migration and transformation, consequently affecting water quality and ecological systems [69, 94]. Within the buffer zone scale, the critical influence ranges for mixed ecological risks extend 2−3 km for cropland and 1−3 km for impervious. Consistent with previous findings, Ma et al. [95] found that organic matter content in water bodies significantly increased with expanding cropland area within 1 km and 2 km buffer zones. Shi et al. [96] conducted a meta-analysis on global landscape changes and water quality, concluding that agricultural land exerts the greatest influence on water quality at the catchment scale and in buffer zones exceeding 2 km. The critical scale range for landscape changes affecting water quality and aquatic ecological risks within watersheds is thus 2 km. Research indicates that buffer zones serve crucial functions in pollution migration, material cycling, and hydrological regulation [97]. Dense buffer zone vegetation captures or slows pollutant transport in runoff, restricting sediment-bound insecticides from reaching off-site surface waters [69, 98]. The relationship between different landscape structures, ecological conditions, and pesticide risks should be carefully considered. Rational land use planning, particularly in buffer zones, combined with the controlled use of highly toxic, persistent pesticides within the turning point range of buffers, can achieve optimal ecological benefits cost-effectively. Sustainable agricultural systems [99] and strategic pesticide use [100], are also key components in this process. Previous studies[39] indicate that maintaining or restoring natural habitats at the periphery of human activity areas reduces pollution risks, while buffer zones can offset production benefits through biodiversity-mediated advantages without compromising crop yields. These findings offer valuable insights for rational buffer zone planning and watershed ecological protection [23, 101].

4.4. Impact of ecological risk thresholds on urban planning and agricultural management

The proportion of land used for human activities, particularly when cropland exceeds 25% or urban areas exceed 30%, substantially influences ecological risks in the context of landscape planning. This corresponds with previous findings, including those of Eduard et al. [102], which demonstrated a positive correlation between agricultural land use and Regulatory Acceptable Concentration exceedances, with the frequency of violations increasing when watershed agricultural land use surpasses 28%. Source management, encompassing pesticide use restrictions and landscape planning, remains paramount in reducing pesticide ecological risks [76]. Voluntarily reducing pesticide use while maintaining agricultural productivity and quality of life proves challenging. Source management should prioritize land use planning to reduce negative impacts on agriculture and production, as land use ratios influence pesticide exposure processes, pollution, decomposition, and transformation, resulting in concerning exposure scenarios [84]. Lower proportions of human land use correlate with reduced ecological risks, potentially linked to landscape complexity. Studies [40] indicate that increased landscape complexity promotes biodiversity and natural pest enemy diversity, leading to reduced pesticide requirements. Land use planning should maintain landscape structural complexity, consistent with Laura et al. [102] findings on functional vegetation diversity, considering richness, evenness, and divergence of functional vegetation to enhance ecosystem resilience against pollution.

Moreover, it's essential to determine critical threshold ranges for agricultural and urban land proportions affecting mixed pesticide risks. Identifying key vulnerable areas and their extent [76], conducting detailed and frequent field surveys to track spatiotemporal variations in pesticide-related environmental risks, and implementing targeted mitigation measures can efficiently address complex ecological issues [16, 103]. The protection of key areas and surface water ecosystems should be maximized through strategic planning of agricultural development patterns and locations. Furthermore, scientifically organizing land use, adjusting the scale of agricultural operations [104], and establishing sustainable agricultural systems [105] represent forward-thinking initiatives. It is recommended to establish a comprehensive national database for future research, categorizing watersheds by crop types, functional vegetation, and land use classifications [76], and to implement online monitoring systems for real-time, high-precision tracking of pesticide application and exposure, thereby effectively mitigating pesticide impacts. The findings of this study offer guidance for future mitigation of aquatic pesticide risks and sustainable agricultural system development.

Urban environmental management requires comprehensive consideration of urban distribution, cropland preservation boundaries, ecological planning, and watershed management. The strategic arrangement of cropland and urban areas is crucial; reducing urban density and agricultural intensification can contribute to more optimal land utilization. According to land use sensitivity thresholds, implementing ecological buffer zones around agricultural lands enables precise control of pesticide pollution, especially buffer zones with functionally diverse vegetation, which more effectively enhance landscape ecological functions. This study's findings offer guidance for alleviating pesticide risks in urban surface waters and advancing the sustainable development of agricultural systems.

4.5. Limitation

The pollution caused by pesticide mixtures represents a global challenge, creating complex toxic effects on non-target organisms through mixture exposure. This study focused solely on additive effects, excluding synergistic and other interactions. While this study examined buffer zone land use impacts on aquatic pesticide ecological risks, future research could explore additional factors such as water catchments, upstream environments, river morphology, soil types, and topography. In addition to total cropland and urban area proportions, future studies could analyze how patch sizes of different land use types in landscape planning influence ecological risks. Notably, this study's sampling was limited to two consecutive seasons and water periods, excluding flood season data. Future research could examine ecological risk responses to land use during periods of heavy rainfall.

5. Conclusions

This research presents an innovative analysis of relationships between multi-scale land use and pesticide ecological risks, uncovering spatial distribution patterns and driving mechanisms in megacity surface waters. The findings demonstrate that urban land use surpasses agricultural land in contributing to pesticide ecological risks within megacities, exhibiting significant spatial scale-dependent effects. The driving factors of pesticide risk highlight the urgent need to address the impact of land use, particularly urban land use, on pesticide exposure. Urban green belts, industrial and construction areas, residential areas, and farmlands all contribute to pesticide exposure and transformation. The identified buffer zone ranges and land use thresholds underscore the positive role of vegetated buffer strips around watersheds in mitigating aquatic pesticide risks and highlight the necessity of rational urban and agricultural planning. To achieve urban sustainability and healthy watershed ecosystem development, land use planning and urban ecological construction can be modified according to urban development goals, economic circumstances, ecological conditions, and natural environmental factors. For instance: (1) expanding forest and grassland coverage within buffer zones surrounding areas of intensive pesticide use and high urban density; (2) employing climate-smart tools [106] to evaluate interactions between climate change and land use, as well as extending buffer zones in regions experiencing intense precipitation and high runoff; (3) following organic agriculture-oriented urban development principles, transitioning selected agricultural lands to organic production while maintaining crop productivity [15]. This research offers quantitative guidance for landscape planning and pesticide risk management.

Future research directions should include: (1) detailed analysis of pesticide sources, transformation processes, and ecological effects in urban environments; (2) development of landscape pattern-based pesticide risk early warning models to inform urban planning decisions; (3) establishment of long-term monitoring networks to evaluate land use change impacts on pesticide risks in the context of climate change.

Glossary

Names	Abbreviations
generalized additive models	GAM
non-parametric change point analysis	nCPA
Neonicotinoid pesticides	NEOs
Organophosphate pesticides	OPPs
Triazine pesticides	TPs
Carbamate pesticides	CAs
Toxicity Units	TU
Predicted no effect concentration	PNEC
Chronic risk thresholds	CRT
Positive matrix factorization	PMF

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Appendix. Supplementary materials

Supplementary material associated with this article can be found.

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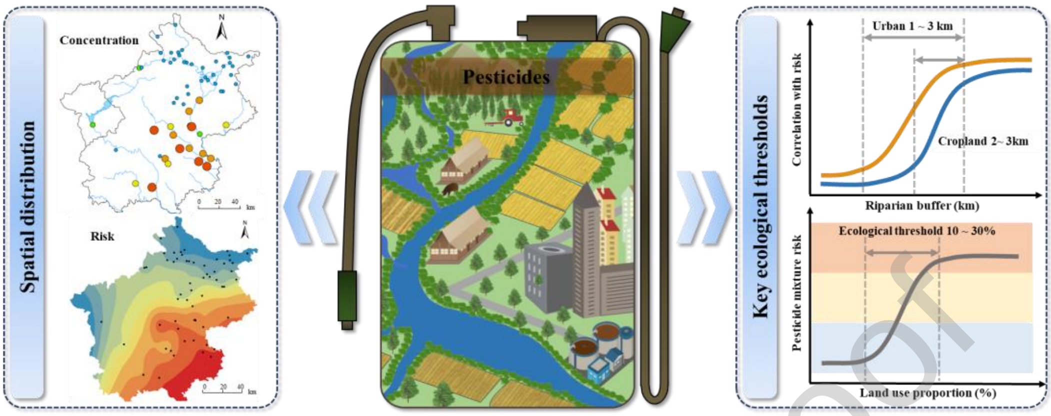
Graphical abstract:



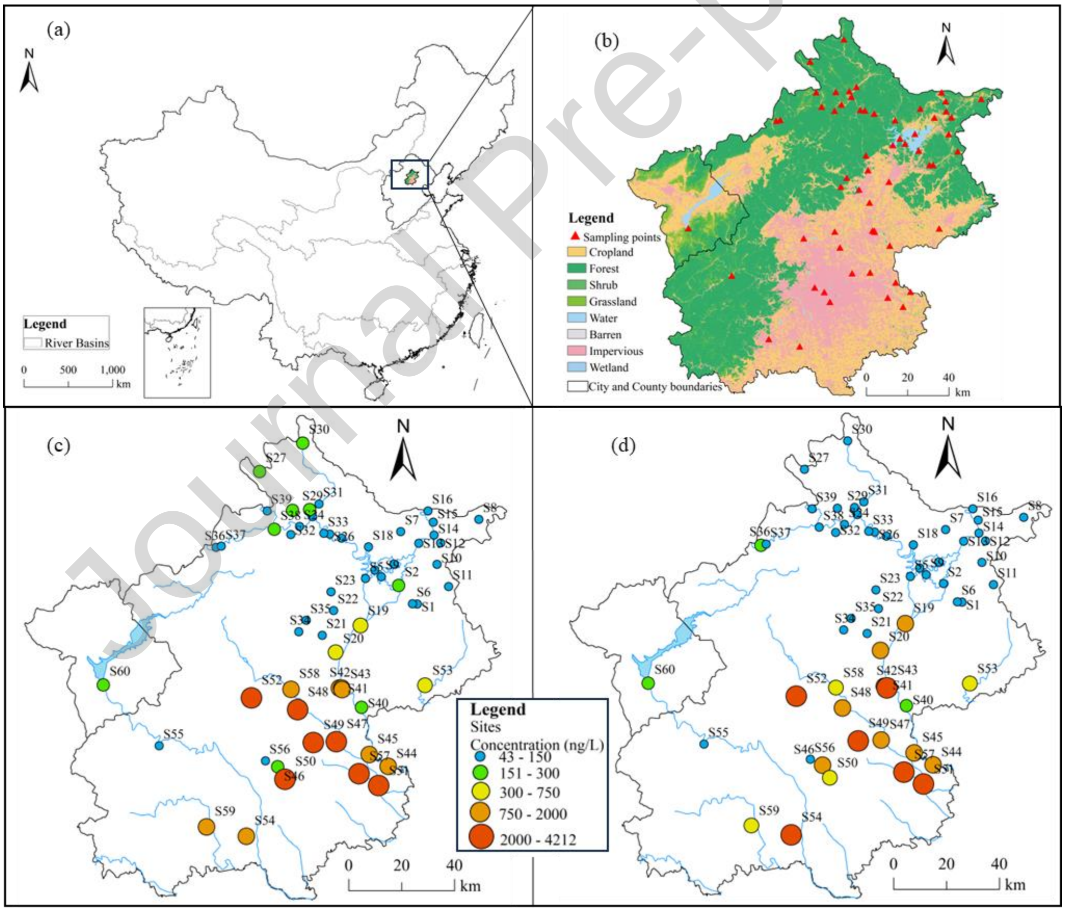


Figure 1 (Color) Sampling locations in Beijing and the spatiotemporal distribution of pesticide concentrations (a) Geographical location of Beijing (b) Distribution of land use types in Beijing (c) Total pesticide concentration in the dry season (d) Total pesticide concentration in the normal season

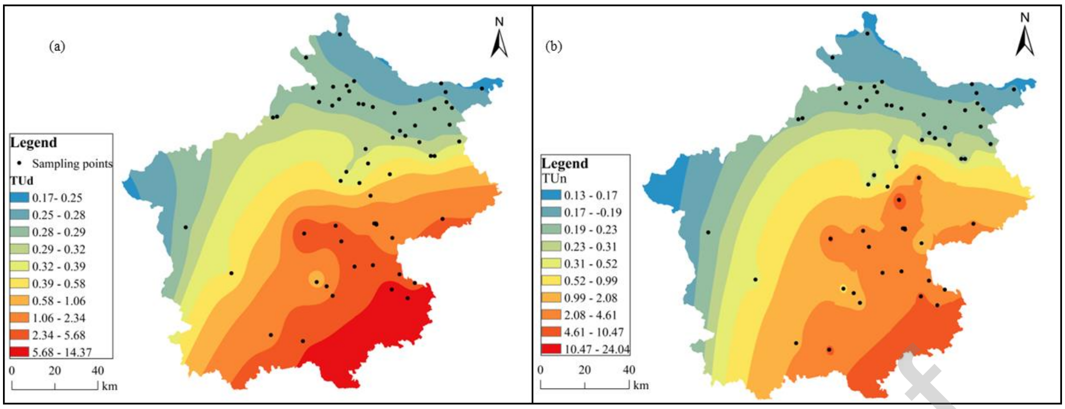


Figure 2 (Color) Characteristics of mixed ecological risk distribution (a) Dry season, (b) Normal season

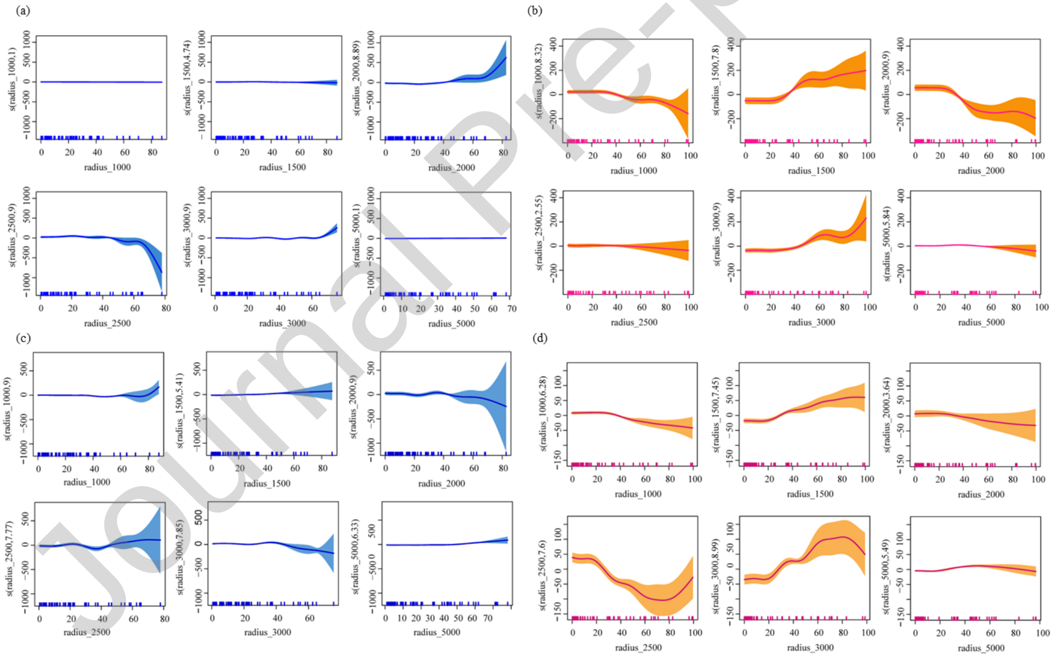


Figure 3 (Color) Residual plots of estimated smoothing functions for land use types and mixed risks in GAM across multiple scales. Dashed lines indicate the 95% confidence intervals for the estimated curves. The vertical bars at the bottom represent the density of data points within the specified range. The vertical axis labels for the smoothing functions include the names of the corresponding explanatory variables and the edf values (effective degrees of freedom), (a) cropland, dry season, (b) impervious, dry season, (c) cropland, normal season, (d) impervious, normal season. For instance, s (radius_1000, 1) denotes the smoothing function for arable land and mixed risk with a 1000 m buffer, where the edf value is 1.

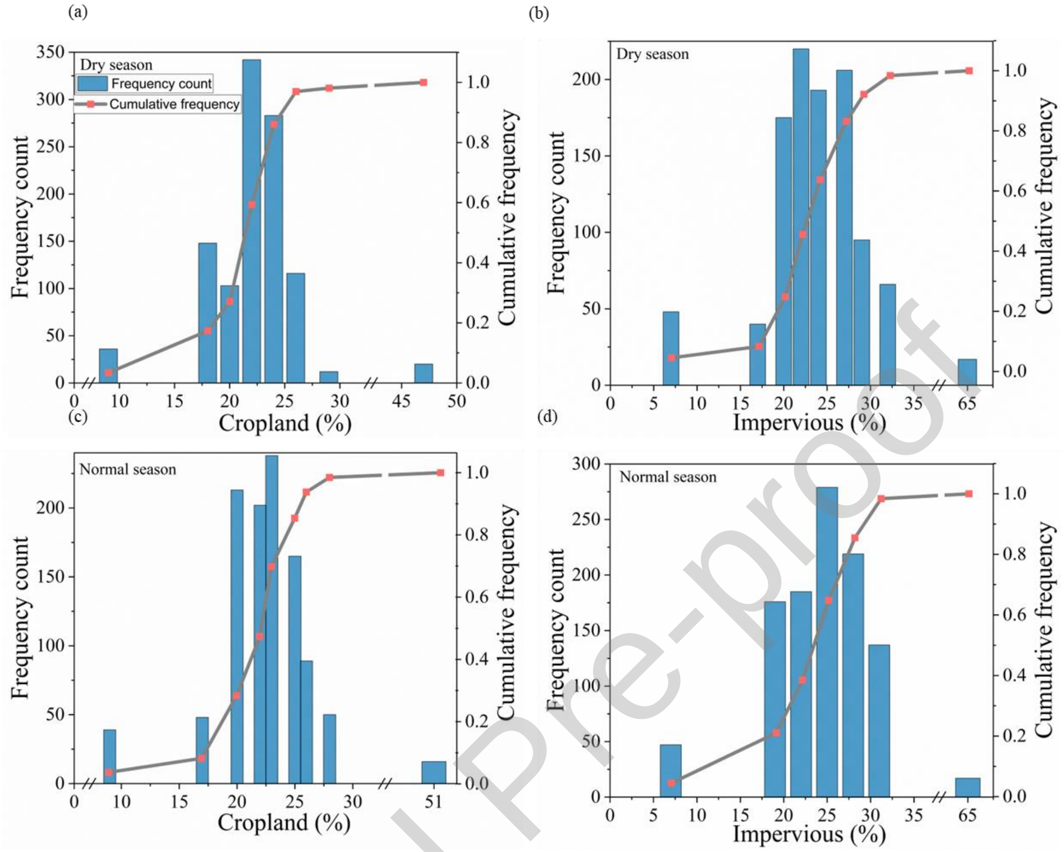


Figure 4 (Color) Frequency counts and cumulative frequencies of change points in mixed risk as related to arable and impervious area proportions. The bar chart illustrates frequency counts, and the dotted line graph represents cumulative frequencies.

Environmental Implication:

This study reveals that urban land use contributes more significantly to pesticide ecological risks than agriculture in megacity surface waters, challenging traditional perspectives. We identified critical land use thresholds (10-25% for cropland, 10-30% for urban areas) and buffer zones (2-3km for cropland, 1-3km for urban areas) where pesticide risks increase dramatically. These quantitative benchmarks provide essential guidance for urban planning and environmental management, demonstrating that landscape optimization can effectively reduce pesticide risks in aquatic ecosystems, offering a novel approach to pollution control in rapidly urbanizing regions worldwide.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Highlights

- Urban land use surpasses agricultural impacts on pesticide risks in megacity waters
- Critical buffer zones (1−3 km) identified for land use effects on pesticide risks
- Land use thresholds (22−25% cropland, 25−30% urban) trigger elevated ecological risks
- Quantitative multi-dimensional model enables landscape-based pesticide management