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Occurrence and risk assessment of current-use pesticides in a tropical drinking water source reservoir in Hainan Province, China†

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The agricultural sector plays a pivotal role in Hainan Province, China; therefore, the utilization of pesticides is indispensable. The current ban on traditional pesticides and ongoing replacement of current-use pesticides (CUPs) have not been accompanied by extensive research on the presence of CUPs in reservoirs, which are vital centralized sources of drinking water. In this study, 26 CUPs was investigated in a drinking water source reservoir, the surrounding watershed, and the surrounding agricultural and domestic discharge water in Hainan Province. The predominant detected CUPs in the study area were clothianidin (CLO), thiamethoxam (THM), acetamiprid (ACE), imidacloprid (IMI), and dichlorvos (DCH). Neonicotinoids (NNIs) were the primary type of pesticide contamination in the study area, with a concentration ranging from not detected (n.d.) to 755 ng L⁻¹ (median of 71.0 ng L⁻¹). The upstream watersheds of the reservoir were primarily contaminated due to agricultural activities, and the highest concentration of individual CUPs, ranging from 102 to 821 ng L⁻¹ (median of 468 ng L⁻¹), was found in agricultural source water. Source identification analysis revealed that the presence of CUPs in the reservoir primarily stemmed from three types of activities: the cultivation of fruit trees around the reservoir, the daily activities of residents, and the agricultural practices in the upstream watershed basin. Risk assessment indicated that DCH, IMI, and THM posed moderate or high risks to aquatic organisms, with an emphasis on the effects of NNIs. The chronic cumulative risk assessment of NNIs was conducted by the relative potency factor approach, and it indicated that infants and young children were the most vulnerable groups and exhibited heightened susceptibility. The potential exposure to NNIs through drinking water was below the recommended relative chronic reference dose, thereby posing no discernible health risks. The results of this study will support the regulation of CUPs in drinking water sources.

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Environmental significance

A comprehensive investigation was conducted to assess the occurrence of 26 current-use pesticides (CUPs) in a drinking water source reservoir, its surrounding watershed, as well as the discharged agricultural and domestic wastewater within Hainan Province. Results revealed that contamination primarily originated from agricultural activities within upstream watersheds, with significantly higher concentrations of individual CUPs detected in agricultural source water. The entry pathways for these CUPs into the reservoir were identified as fruit tree cultivation near the reservoir area, daily human activities, and agriculture practices within the upstream watershed basin. These findings offer valuable insights for formulating effective regulatory measures aimed at managing CUP contamination risks associated with drinking water sources.

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1. Introduction

Hainan Province, located in the tropics of China, has abundant agricultural resources that constitute a vital sector of its economy. Moreover, the prevalence of pests, diseases, and weeds in tropical regions necessitates the high usage of pesticides in this province.¹ However, the majority of the used pesticides do not target crops, instead, they persist in various environmental media and are commonly detectable in soil^{2–4} and surface waters.^{5–7} Pesticide residues can be introduced into rivers and reservoirs *via* the upstream precipitation and



agricultural practices.^{8,9} Therefore, pesticide residues in drinking water sources can pose a significant threat to the ecosystem, necessitating urgent attention and action.

Based on their properties, pesticides can be classified into several major groups, including organochlorine pesticides (OCPs), organophosphorus pesticides (OPPs), and the recently prominent neonicotinoid pesticides (NNIs).¹⁰ Most OCPs are categorized as persistent organic pollutants (POPs) and are prohibited globally because of their high toxicity.¹¹ However, certain OCPs, such as aldrin, are still used in China.¹² Subsequent studies have demonstrated the detrimental impacts of OPPs on both human and animal health, leading to the prohibition of certain OPPs in agricultural practices.^{12,13} Therefore, in recent years, the traditional pesticides characterized by high toxicity and persistence have been widely replaced by low-toxicity alternatives such as neonicotinoids.¹⁴ Currently, NNIs occupy nearly a quarter of the insecticide market and have experienced continuous sales growth in recent years.¹⁵ Research has shown that NNI exposure poses a threat to aquatic biodiversity,¹⁶ and the potential human health risks associated with NNI contamination have recently garnered remarkable attention.

Drinking water sources play a vital role in the regional water environment, as they serve as the crucial resources for urban water supply.¹⁷ The formation of reservoirs through surface runoff can lead to the pollutant accumulation, transforming drinking water sources into the potential pesticide repositories¹⁸ and thereby causing harm to plants, animals, and humans. The studies of pesticides in large-scale drinking water sources, have primarily focused on the impacts of traditional pesticide pollutants.^{9,19} However, few studies have examined the effects of multiple conventional and alternative pesticides on drinking water sources.²⁰

A previous study indicated that the contamination of the Tighra reservoir in India was dominantly attributed to the utilization of pesticides in nearby agricultural fields.²¹ Human activities have significantly influenced the environment, as evidenced by the elevated levels of OCPs observed in the upstream area of the Miyun Reservoir, which is predominantly characterized by cornfields and orchards.³ Furthermore, herbicides have been found to pose a serve threat to the aquatic organisms in the tributaries and the reservoir itself.²² Additionally, NNIs are extensively used pesticides, and their presence in the rivers and lakes in recent years has been the focus of various studies.^{23,24} However, research on their presence in drinking water sources are limited.^{25–27}

The upstream or vicinity of the drinking water source reservoir in Hainan Province is commonly characterized by frequent agricultural production activities. Furthermore, given the previous studies of the pesticide contamination in the drinking water sources primarily focused on individual traditional pesticides,¹⁹ necessitate imminent investigation into multiple pesticides. High annual precipitation in the region facilitates the migration of pesticides from the terrestrial environment to the aquatic ecosystems, and their accumulation in the reservoirs. Therefore, it is imperative to deepen the understanding of the pollution characteristics and the potential risks

associated with multiple pesticides in the drinking water sources.²⁸

The aim of this study was to investigate the abundance, composition, and spatial distribution of multiple current-use pesticides (CUPs) in a drinking water source reservoir and its surrounding agricultural sources, domestic sources, and watersheds in Hainan Province. Furthermore, the sources of CUPs in the water environment were identified, and the potential risks to aquatic ecosystems and humans from pesticide contamination were assessed in the drinking water sources. The findings of this study will contribute to a comprehensive understanding of the occurrence and fate of CUPs in drinking water sources and their surrounding aquatic environments, thereby providing a theoretical foundation for evaluating levels of CUP contamination in reservoirs.

2. Materials and methods

2.1 Study area and sample collection

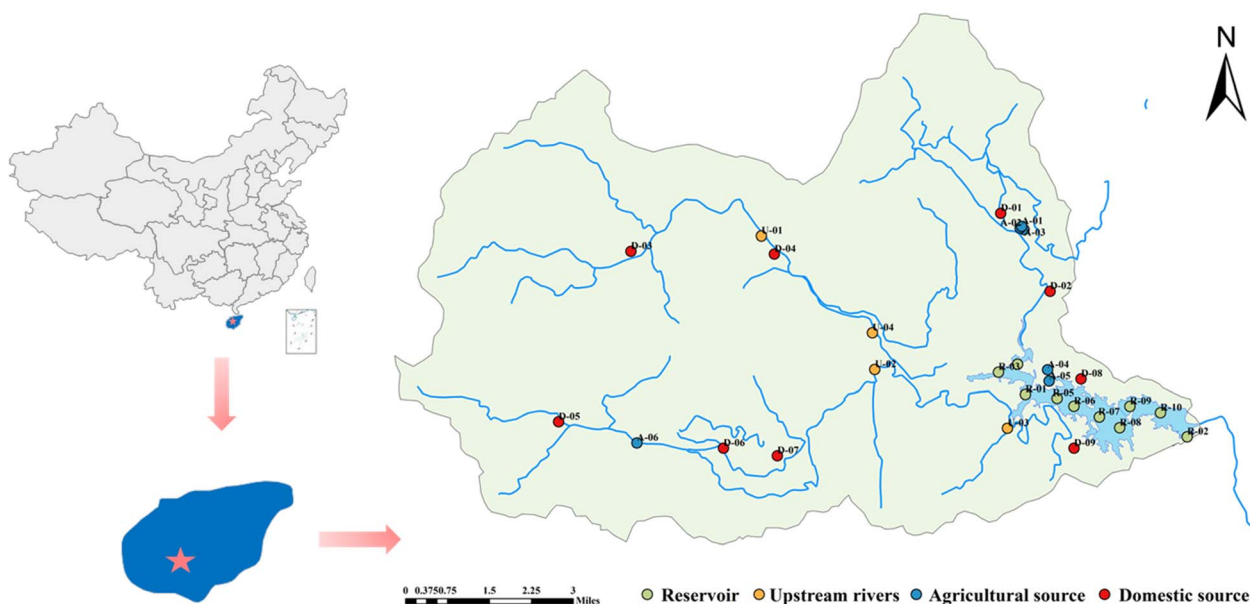
The study area serves as a crucial centralized urban drinking water source reservoir in Hainan Province, China (Fig. 1), which provides a daily supply of approximately 220 000 t of water. Surface water samples were collected from the reservoirs ($n = 10$), the upstream rivers ($n = 4$), the surrounding agricultural sources ($n = 6$), and the domestic sources ($n = 9$). Surface water samples were collected in May 2023. Agricultural sources refer to the surface water in cultivation area catchments with mangoes, longans, betel nuts, and other commercial crops located in the upstream of the reservoirs, whereas domestic sources refer to the effluent from the domestic wastewater treatment plants in the residential areas around the reservoirs. In each site, 1 L of surface water was collected and stored in a brown glass bottle. Afterward, 50 mL of methanol (MeOH) was added to each sample to inhibit microbial growth. All water samples were placed in the refrigerator (4 °C) for chilling within 48 h after the collection.

2.2 Chemicals and reagents

The target CUPs include 9 NNIs, 7 OPPs, and 10 other pesticides (OthPs; herbicides, fungicides, *etc.*). Dinotefuran (DNT), acetamiprid (ACE), nitenpyram (NTP), clothianidin (CLO), thiacloprid (THA), thiamethoxam (THM), imidacloprid (IMI), imidaclothiz (IMT), flonicamid (FLO), phoxim (PXM), dichlorvos (DCH), malathion (MLT), dimethoate (DMT), trichlorfon (TCF), phorate (PHT), chlorpyrifos (CPF), atrazine (ATR), butachlor (BTC), acetochlor (ATC), metalaxyl (MLX), carbendazim (CBZ), thiophanate-methyl (TPH), pyraclostrobin (PCT), spirodiclofen (SPD), indoxacarb (IDC), and emamectin benzoate (EMB) were purchased from First Standard (China). Four isotope-labeled internal standards (ISs), namely carbendazim- d_3 (CBZ- d_3), acetamiprid- d_3 (ACE- d_3), clothianidin- d_3 (CLO- d_3), and imidacloprid- d_3 (IMD- d_3), were also purchased from First Standard (China).

All other chemicals were of HPLC grade. Detailed information is provided in the ESI† (Text S1 and Table S1).





2.3 Sample pre-treatment and instrumental analysis

Target chemicals were analyzed using a high-performance liquid chromatography-triple quadrupole mass spectrometer (UPLC-MS/MS; Waters, TQ-S Micro). The chromatographic and MS parameters are described in detail in the ESI[†] (Text S2).

One program blank sample (replaced with Milli-Q water) was analyzed every 10 samples to assess the potential contamination, and the average levels of individual target chemicals detected in the program blanks ($n = 5$) were subtracted from the water samples. Additionally, 3 spiked blank samples were pre-treated and analyzed using the same procedures as the water samples. The accuracy was calculated as the recoveries of analytes in the natural spiked samples, *i.e.*, 53.0–111.0%.

2.5 Risk assessment

The aquatic risk assessment for all detected CUPs was conducted using a risk quotient (RQ) approach, which involved comparing the measured environmental concentration (MEC) to the predicted no-effect concentration (PNEC) (eqn (1)).²⁹

$$RQ = MEC/PNEC \quad (1)$$

The no-observed-effect concentration (NOEC) values (or in their absence, EC₅₀ values) were used to calculate the PNEC values. To mitigate the uncertainties associated with assessment, including accuracy, inherent variability, model errors, and insufficient toxicity data, the PNEC values were estimated by dividing the lowest NOEC or EC₅₀ value of the most sensitive species by an appropriate safety factor (AF) for each trophic level—fish, algae, and aquatic invertebrates.¹⁰ Hazard quotient data for risk assessment of the same pollutant may vary across different toxicological parameters. To avoid underestimating the existing ecological risks, we selected the lowest PNEC from the NORMAN Ecotoxicology Database as the input value, which are presented in Table S4† within this study.³⁰ The restrictive RQ ranking is determined as follows: a value of $0 \leq RQ < 0.1$ indicates a low risk level; a value of $0.1 \leq RQ < 1$ indicates a medium risk level; and an RQ value ≥ 1 indicates a high risk level.³¹

2.6 Health risks

The chronic reference dose (cRfD) of each NNI and the corresponding relative potency factor (RPF) were normalized to the

IMI,³² as shown in Table S5.† We utilized the RPF method to assess the chronic cumulative risk of four NNIs (ACE, CLO, THM, and IMI) in the reservoir water samples.

IMI_{eq.} was calculated as follows:

$$\begin{aligned} \text{IMI}_{\text{eq.}} &= \sum_i \text{NNI}_i \times \text{RPF}_i \\ &= \text{IMI} \times 1 + \text{THM} \times 9.5 + \text{ACE} \times 0.803 \\ &\quad + \text{CLO} \times 5.816 + \text{DNT} \times 2.85 \end{aligned} \quad (2)$$

where IMI_{eq.} is the total IMI-equivalent NEO concentration. The pesticide residues in the study area, which serves as a crucial drinking water source for the city, pose a potential threat to human health. However, the effectiveness of conventional drinking water treatment processes is limited in terms of pesticide removal efficiency.³³ We estimated the daily intake (EDI; ng kg⁻¹ bw per d) of NNI in the reservoir, as follows:

$$\text{EDI} = \text{IMI}_{\text{eq.}} \times \text{DIR} \times \text{AR} \quad (3)$$

where DIR represents the daily water ingestion rate (L per kg bw per day) (Table S6†), while AR denotes the absorption rate of a human, which remains constant at 100%.³⁴

2.7 Statistical analysis

The statistical analysis was conducted utilizing the SPSS software (version 22.0; SPSS Inc., Chicago, IL, USA) for the analytes

with a detection frequency (DF) > 50%. The normality of all data was assessed prior to the statistical analysis; non-parametric tests were employed for data that did not exhibit a normal distribution, and to examine the correlation between the concentrations of different compounds, a significance criterion of $P < 0.05$ was applied. For the statistical analysis, the chemical concentrations below the MQL were replaced with 1/2 MQL. Principal component analysis (PCA) was performed to evaluate the potential sources of CUPs in water.

3. Results and discussion

3.1 Levels of CUPs in surface water

The DFs and concentrations of CUPs are listed in Table S7.† Among the 26 targeted chemicals, 9 NNI, 4 OPPs, and 10 OthPs were present in water samples, with concentrations of n.d.–755 ng L⁻¹ (median of 71.0 ng L⁻¹), n.d.–297 ng L⁻¹ (median of 3.91 ng L⁻¹), and n.d.–439 ng L⁻¹ (median of 28.3 ng L⁻¹), respectively (Fig. S1†). NNIs were the primary CUPs in the study area, and their relative contribution (62.7%) was significantly higher than OthPs (27.3%) and OPPs (9.98%) (Fig. S2†).

The main NNIs in surface water samples were CLO, THM, ACE, and IMI, with DFs of 89.7%, 75.9%, 72.4%, and 65.5%, respectively (Table S7 and Fig. S3†). CLO, THM, and IMI have also been detected predominantly in groundwater in Iowa, USA³⁵ and the Yangtze River, China 14. In this study, DNT was detected at only one sampling site in the agricultural source (A-

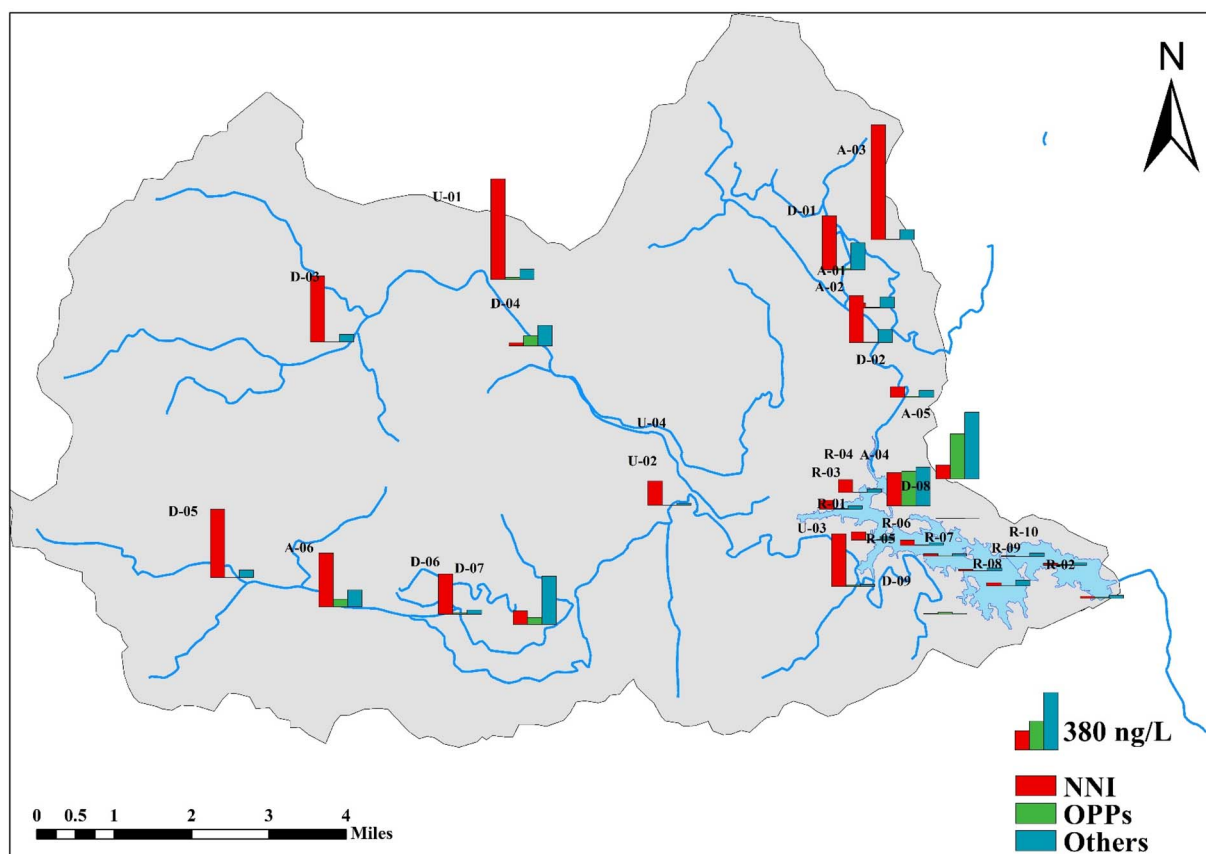


Fig. 2 Histogram of the spatial distribution of NNI, OPPs, and OthPs in the study area.



Table 1 Concentrations (ng L⁻¹) of pesticides in surface water in the reservoirs (*n* = 10), upstream watersheds (*n* = 4), agricultural sources (*n* = 4), and domestic sources (*n* = 9)

Compounds	Reservoirs				Upstream watersheds				Agricultural sources				Domestic sources			
	Range ^a	Median	Average	DF ^b (%)	Range	Median	Average	DF (%)	Range	Median	Average	DF (%)	Range	Median	Average	DF (%)
DNT	n.d. ^c	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.-2.07	n.d.	0.35	0	n.d.	n.d.	n.d.	0
ACE	n.d.-0.56	0.37	0.28	60	n.d.-13.7	5.18	6.01	75	n.d.-95	9.63	22	66.7	n.d.-21.9	2.46	4.62	77.8
NTP	n.d.-n.d.	n.d.	n.d.	n.d.	n.d.-0.49	n.d.	0.12	25	n.d.-0.49	n.d.	0.08	16.7	n.d.	n.d.	n.d.	n.d.
CLO	2.7-16.2	6.93	8.12	100	n.d.-102	82.4	66.7	75	n.d.-210	69.1	80	66.7	n.d.-91.2	11.9	25.5	88.9
THA	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	n.d.	n.d.-0.39	n.d.	0.043	11.1
THM	n.d.-60.3	10.8	20.3	90	n.d.-465	132	182	75	n.d.-539	15.2	144	66.7	n.d.-333	45.1	132	66.7
IMI	n.d.-6.94	n.d.	1.92	30	n.d.-96	26.1	37.1	75	n.d.-210	12.4	46.6	66.7	n.d.-92.9	17.8	25.9	88.9
IMIT	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	0	n.d.-1.4	n.d.	0.156	11.1
FLO	n.d.-0.70	n.d.	0.193	30	n.d.-0.93	0.26	0.37	50	n.d.-2.04	n.d.	0.53	16.7	n.d.-0.54	n.d.	0.0596	11.1
∑ ₁₀ NNI	3.38-84	17.7	30.8	100	n.d.-662	254	292	75	31.6-755	264	293	83.3	3.83-448	89.2	188	100
PXM	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	0
DCH	n.d.-6.05	2.41	2.42	70	n.d.-6.05	3.27	3.15	75	n.d.-286	4.17	84.1	66.7	n.d.-48.1	4.08	9.09	77.8
MLT	n.d.	n.d.	n.d.	n.d.	n.d.-0.55	n.d.	0.14	25	n.d.-16.3	0.28	4.2	66.7	n.d.-8.64	0.155	1.39	55.6
DMT	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	n.d.
TCF	n.d.	n.d.	n.d.	n.d.	n.d.-4.15	1.62	1.85	50	n.d.-46.9	0.78	8.5	33.3	n.d.-6.48	n.d.	0.72	11.1
PHT	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	0
CPF	n.d.	n.d.	n.d.	n.d.	n.d.-3.61	0.88	1.34	50	n.d.-1.96	n.d.	0.63	33.3	n.d.-32	n.d.	5.46	33.3
∑ ₇ OPPs	n.d.-6.05	2.41	2.42	70	n.d.-14.4	5.77	6.48	75	2.1-297	27.9	97.4	83.3	n.d.-65.4	7.47	16.7	77.8
ATR	n.d.-1.39	n.d.	0.23	30	n.d.	n.d.	n.d.	0	n.d.-1.3	n.d.	0.22	16.7	n.d.-0.61	n.d.	0.068	11.1
BTC	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	0
ATC	n.d.	n.d.	n.d.	n.d.	n.d.-11.8	n.d.	2.94	25	n.d.-55.4	n.d.	9.23	16.7	n.d.-27.5	n.d.	3.06	11.1
MLX	10.2-21.1	12.9	13.8	100	n.d.-42.5	4.34	12.8	75	3.1-69.9	22.5	29.6	83.3	n.d.-36.8	6.99	12.5	88.9
CBZ	3.53-12.6	5.62	6.24	100	n.d.-6.79	5.2	4.3	75	n.d.-322	17.1	66.6	66.7	n.d.-139	8.93	32.2	77.8
TPH	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	0	n.d.-6.14	n.d.	1.62	33.3
PCT	n.d.	n.d.	n.d.	n.d.	n.d.-7.65	1.39	2.61	75	0.177-63.7	1.78	13.5	83.3	n.d.-4.23	1.6	1.5	66.7
SPD	n.d.-2.9	1.09	1.28	50	n.d.-2.8	1.14	1.27	50	n.d.-245	12.4	51.9	66.7	n.d.-202	n.d.	37.9	33.3
IDC	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	0	n.d.-6.84	n.d.	0.76	11.1
EMB	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	0	n.d.-5.88	n.d.	0.653	11.1
∑ ₁₀ OthPs	14.9-36.7	20.4	21.5	100	n.d.-68.7	13.5	23.9	75	66.4-439	98.1	171	83.3	n.d.-317	50.4	90.3	88.9
∑ ₂₆ CUPs	25.7-108	43.6	52.4	100	n.d.-730	267	316	75	102-821	468	464	83.3	3.83-533	293	278	100

^a Range: minimum-maximum. ^b DF: detection frequency. ^c n.d.: not detected.

06), which could be related to the fact that DNT is mainly applied in rice cultivation.³⁶ In contrast, the study area does not engage in rice cultivation.

DCH was the predominant OPP in this study, with a DF of 75.9%, whereas MLT, TCF, and CPF had DFs <50% (Table S7†). The concentrations of OPPs ranged from n.d. to 297 ng L⁻¹ (median of 3.91 ng L⁻¹), with the highest value was observed at the agricultural source sites, likely due to the severe contamination of utilizing DCH (Fig. S4†).

OthPs were mainly represented by the fungicides, MLX and CBZ, with DFs of 93.1% and 86.2%, respectively. MLX and CBZ concentrations were n.d.–69.9 ng L⁻¹ (median of 11.4 ng L⁻¹) and n.d.–322 ng L⁻¹ (median of 5.71 ng L⁻¹), respectively (Table S7 and Fig. S3†). MLX has been widely detected in previous studies^{37,38} because of its stable structure and long half-life (115 d at pH 9).³⁹ CBZ is a common fungicide, and the high concentration of CBZ in this study may be related to its high consumption and wide range of uses. However, the concentrations of CBZ in our study area were lower than the Huangpu River, China (41.9–607.0 ng L⁻¹),⁴⁰ and higher than the Vistonis Lake, Greece (<25 ng L⁻¹).⁴¹

3.2 Spatial distribution and source identification

3.2.1 Spatial distribution. As shown in Fig. 2, all types of sampling sites in the upstream of the reservoir were generally contaminated with CUPs. Pesticide levels were usually low at the sampling sites in the reservoir, with $\sum$ CUPs of 25.7–108.0 ng L⁻¹ (median of 43.6 ng L⁻¹) (Table 1 and Fig. 2). $\sum$ CUPs showed the highest concentrations (102–821 ng L⁻¹; median of 468 ng L⁻¹) in the surface runoff of the agricultural source sites (Table 1). The detection of numerous compounds from diverse categories primarily occurred at the agricultural source sites in relation to the pesticide contamination—especially at sites A-04 (mango cultivation) and A-05 (longan cultivation). This phenomenon can be attributed to the cultivation practices of fruit crops, and the specific pesticides employed during different periods,⁴² and the persistent historical use of pesticides at this particular site. Previous studies have demonstrated that the primary pathway for the non-point emissions from pesticide land use is the surface runoff into streams.^{15,43} Therefore, it is crucial to prioritize regulating pesticide use in the agricultural developments near the watersheds.

NNIs were widely detected in the agricultural sources. The highest value, 755 ng L⁻¹, was found at the agricultural source at site A-03 (Table 1). This site is part of the rice–vegetable crop rotation, where NNIs are commonly utilized, and THM and CLO are the dominant chemicals. Additionally, A-02 is mainly planted with betel nuts, and the NNI concentration there was 310 ng L⁻¹. This suggested that these neonicotinoids from the agricultural sources in the study area may have contaminated the surface water through the surface runoff (Fig. 1).

Among the upstream section sites, NNIs were absent only at site U-04, which is located downstream of site U-01. It is likely that the degradation of NNIs occurred during the discharge process downstream,⁴⁴ resulting in the absence of NNI at this particular site. However, in the other three upstream sampling

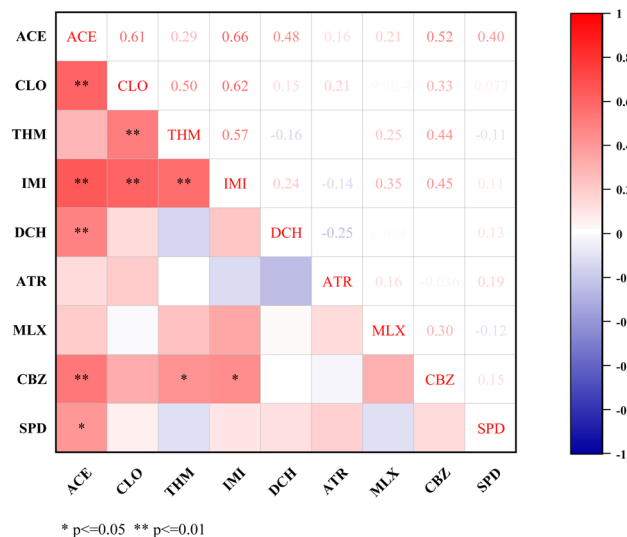


Fig. 3 Correlation between the nine predominant pesticides detected in water (** Correlation (Corr.) is significant at the 0.01 level (2-tailed). * Corr. is significant at the 0.05 level).

sites (U-01, U-02, and U-03), $\sum$ NNIs were in the range of 160–662 ng L⁻¹, with IMI, THM, and CLO being the predominantly chemicals (Fig. 3). Similarly, these CUPs were also the main NNIs in the domestic and the agricultural sources (Table 1). It has been shown that the activities in agricultural and urban areas are the primary sources of NNIs in the water environment.⁴⁵ The NNI pollution in the watersheds in this study should also be closely related to human activities.

OPP were mainly found at the sampling sites A-04 (mango cultivation) and A-05 (longan cultivation), with DCH being the predominant chemical (Fig. 4). According to the latest report from the China Pesticide Information Network, despite being classified as a group 2B carcinogen by the World Health Organization in 2017 (<http://www.chinapesticide.org.cn>), DCH continues to be utilized in China because of its effective insecticidal properties, prompt decomposition rate, and relatively brief residual duration. Of the 7 OPPs, only DCH was detected in the reservoirs, albeit at a low concentration (n.d.–6.05 ng L⁻¹; mean of 2.42 ng L⁻¹)—lower than the average level (17.8 ng L⁻¹) in surface water of China in a previous study.⁴⁶

The most prominent sites for OthP concentrations, namely sites A-04 (254 ng L⁻¹), A-05 (439 ng L⁻¹), and site D-07 (317 ng L⁻¹) for the domestic sources, were included in the dataset (Fig. 2). This could be attributed to the transition of agricultural cultivation within the residential areas of this locality, as indicated by the increase of vegetable acreage in recent years (Fig. 2). The OthPs at site D-07 were mainly SPD and MLX.

The limited presence of CUPs in the reservoir sites may be attributed to the transfer or the degradation of CUPs from upstream watersheds before reaching the reservoirs coupled with their dilution through the continuous confluence of the tributary streams.⁴⁷



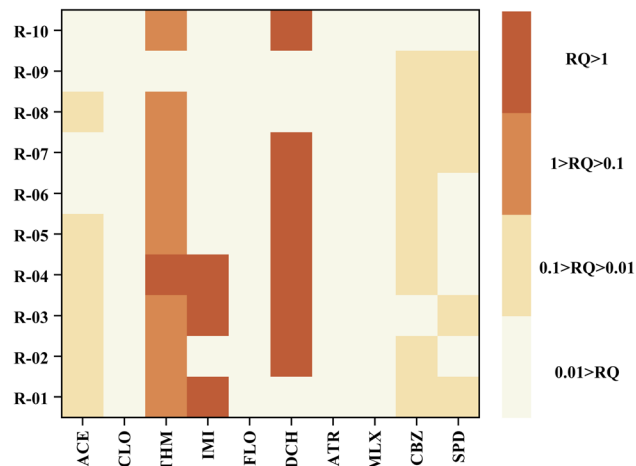


Fig. 4 Principal component analysis results based on the concentrations of NNI, OPPs, and Others (PC1, 32.1% variance; PC2, 23.5% variance). The figure legends represent the factor loadings (a) and factor scores (b).

The heatmap (Fig. 4a) demonstrated the significant positive correlations between IMI and ACE, CLO, and THM ($P < 0.01$), suggesting the potential synergistic utilization of IMI and the other three NNIs in the study area. The PCA plot showed that the first two principal components accounted for 32.1% and 23.5% of the total variance, respectively. CBZ, ATR, SPD, DCH, and ACE in the water were the main contributors to PC1, whereas IMI, THM, CLO, and MLX were the main contributors to PC2 (Fig. 5). A disparity in the origin of the two factors may exist: Factor 1 primarily comprises widely employed fungicides, herbicides, insecticides, acaricides and the only one

The factor score plot (Fig. 4b) indicated that the agricultural sources are distributed across both Factors 1 and 2. In contrast, Factor 1 is primarily associated with the agricultural source sites A-04 and A-05, which are near the reservoir perimeter and encompass extensive areas of fruit tree cultivation (mango and longan). Conversely, Factor 2 was closely linked to various agricultural and domestic sources, including the betel nut planting, the rubber planting, and the rice-vegetable crop rotation. Furthermore, the domestic source sites contributed to the watershed through the discharge of treated domestic wastewater. These findings demonstrated that the pesticide sources in Factor 1 were predominantly influenced by fruit tree planting around the reservoir, whereas pesticides from domestic and agricultural sources dominated Factor 2. These findings suggest that multiple factors drive pesticide pollution in the reservoir. The primary pesticide sources in drinking water reservoirs are associated with the following three types of pesticide usage: (i) the cultivation of fruit trees planted near the reservoir, (ii) the application by residents in residential areas, and (iii) the agricultural practices in the upstream watersheds. This comprehensive analysis underscores the critical need for developing targeted management strategies to address agricultural and domestic sources in order to mitigate pesticide pollution around water reservoirs.

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