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Research Article

Post-ban Assessment of Residual Paraquat in Surface Water and Self-Reported Health Symptoms in An Intensive Agricultural Watershed, Thailand

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Abstract

Despite Thailand's nationwide ban on paraquat, its strong adsorption to soil particles contributes to its long-term persistence in agricultural landscapes. While the ban has curtailed new applications, evidence evaluating environmental residues in postban settings remains limited. In this study, surface water quality indicators and paraquat residue across three districts in Phitsanulok Province were assessed using integrated environmental monitoring, ecological risk characterization, chronic and upper-bound screening hazard quotient (HQ) analysis, and a household questionnaire of 231 residents in the district with the highest paraquat detection frequency and screening-level HQs. The results indicated that paraquat was detected at 75% of the sampling sites in Phrom Phiram, whose mean concentration was the highest (0.39 ± 0.06 mg L^{-1}). Phosphate levels were moderately positively correlated with paraquat (r = 0.426, p = 0.030), indicating an association between the two variables. The screening-level HQs exceeded the ecological benchmark in all the districts under the evaluated exposure scenarios, including the upper-bound screening scenario. Among the surveyed farmers, 69% reported acute symptoms following herbicide exposure, primarily ocular and respiratory irritation. The persistence of paraquat residues indicates potential ecological concern, whereas the high prevalence of self-reported symptoms highlights broader occupational health concerns among farmers. These findings underscore that regulatory bans should be complemented by ongoing environmental monitoring and management to mitigate residual exposure risks.

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Introduction

The legacy of agrochemical contamination remains a significant public health challenge, particularly in countries with intensive agricultural practices, where herbicide residues can be mobilized through hydrological transport pathways, leading to the contamination of aquatic systems (Nguyen et al., 2018). Owing to the documented risks to human and ecological health, including potential carcinogenic effects, paraquat has been prohibited or subjected to strict regulatory control in several regions, including the European Union and parts of Asia (Carrillo et al., 2025; Donaher and Van den Hurk, 2023; Konthonbut et al., 2018).

In Thailand, paraquat was widely used for agricultural weed control prior to its prohibition (Konthonbut et al., 2018; Srisuma et al., 2023). Despite its agronomic benefits, paraquat has raised substantial human health and environmental concerns (Laohaudomchok et al., 2021). Acute paraquat exposure can result in severe and potentially fatal poisoning. Long-term paraquat

exposure has also been associated with an increased risk of Parkinson's disease (Sharma and Mittal, 2024). Paraquat also poses ecological concerns in aquatic environments, particularly for aquatic primary producers (Donaher and Van den Hurk, 2023). Furthermore, research conducted prior to the ban documented paraquat exposure among vulnerable populations, including pregnant women and neonates in agricultural communities (Konthonbut et al., 2018). In Thailand, paraquat was reclassified as a Type 4 hazardous substance under the Hazardous Substances Act, resulting in a nationwide prohibition effective from 1 June 2020 through the Notification of the Ministry of Industry (Ministry of Industry, 2020). Notably, despite the ban, its environmental residues remain a critical concern, particularly regarding the potential for continued water contamination and its long-term effects on vulnerable populations. Consequently, monitoring and understanding the environmental fate of paraquat following regulatory prohibition is essential for assessing residual risks; however, an integrated framework that multidimensionally links physical water quality variables, localized ecological hazard indices, and self-reported symptom profiles at the community level remains limited. Understanding these interconnected parameters is critical for designing target-specific postregulation management strategies.

Paraquat strongly adsorbs to soil particles, particularly clay and organic matter, contributing to its prolonged persistence in soils (Flafel et al., 2024). In aquatic environments, paraquat may be associated with sediments and suspended particles, potentially contributing to its retention and persistence within these systems (Flafel et al., 2024). Such contamination may pose ecological risks, particularly to aquatic primary producers (Donaher and Van den Hurk, 2023). The interaction of paraquat with soil organic matter may further reduce its bioavailability and contribute to its long-term retention in soil (Flafel et al., 2024). Hydrological conditions may influence the transport and distribution of paraquat residues in surface waters (Nguyen et al., 2018). The strong adsorption of paraquat to soil particles allows residues to remain associated with eroded sediments, which may facilitate their lateral transport into adjacent surface-water systems (Flafel et al., 2024).

Surface water quality parameters, including pH, turbidity, total dissolved solids (TDS), dissolved oxygen (DO), and total phosphorus/phosphate, provide complementary information for characterizing environmental conditions in aquatic systems where paraquat residues may occur (Ikpesu, 2015). The inclusion of nutrient-related parameters is also relevant in agricultural settings, where paraquat residue may occur in nutrient-containing runoff (Udomkun et al., 2025). Phitsanulok Province, located in Thailand's lower northern region, is a predominantly agricultural area, with rice cultivation representing an important component of agricultural land use (Sornlorm et al., 2022). Paraquat was widely used for agricultural weed control in Thailand prior to its prohibition (Laohaudomchok et al., 2021). Previous studies have identified paraquat exposure among agricultural workers, with exposure-related concerns associated with pesticide handling practices and limited use of personal protective measures (Laohaudomchok et al., 2021; Srisuma et al., 2023). The Nan River watershed, which flows through the province, serves as an important water resource for the region. Given the intensive agricultural activities within the study area, surface-water systems may be susceptible to agro-chemical inputs through hydrological runoff pathways (Nguyen et al., 2018).

To address these environmental and public health concerns, this study focuses on agricultural watershed areas with diverse historical herbicide use patterns and adopts an integrated assessment approach. Environmental monitoring of paraquat residues and key surface-water quality indicators was combined with a structured questionnaire survey to assess herbicide use practices and document self-reported health symptoms. By integrating environmental residue monitoring, ecological risk screening using hazard quotient (HQ) estimates, and self-reported health information, this study aims to provide complementary evidence on environmental contamination and broader agricultural health concerns in a postban setting.

Materials and methods

1) Study area and design

Water samples were collected from October to December 2024 in three districts of Phitsanulok Province, Thailand: Phrom Phiram, Wang Thong, and Mueang. This period was selected to capture the hydrological transition from the late wet season to the early dry season, as seasonal hydrological conditions may influence the occurrence and distribution of paraquat in surface waters (Ikpesu, 2015; Nguyen et al., 2018). During this period, hydrological processes may facilitate the transport of particle-associated paraquat residues from agricultural soils to adjacent surface waters (Flafel et al., 2024; Nguyen et al., 2018). Accordingly, the sampling period was intended to characterize conditions during this seasonal transition rather than to represent year-round paraquat concentrations, which may vary across monsoonal, dry-season, and irrigation periods. The sampling stations were strategically positioned on the basis of hydrological flow dynamics and the density of adjacent agricultural land use to capture spatial contrasts in surface-water conditions across areas with differing agricultural intensities (Figure 1).

Phrom Phiram District is predominantly agricultural, especially known for rice cultivation, with an extensive

network of irrigation canals sourced from the upstream area of the Nan River watershed. Eight sampling stations were established across this area to characterize surface-water conditions in an intensive rice-growing setting. The selection of this agricultural setting was supported by previous evidence demonstrating spatial associations between rice-field land use and pesticide-related health patterns in Thailand, with herbicide use identified as an important agricultural factor in rice-growing areas (Sornlorm et al., 2022). Phitsanulok was selected on the basis of reports of paraquat residues in surface water, as highlighted in a recent review of the situation in Thailand (Laohaudomchok et al., 2021).

Two comparison areas with contrasting land use characteristics were selected for spatial comparison. The selection of contrasting land use settings was based on the general premise that the occurrence and distribution of contaminants may vary across different land use types (Manamsa et al., 2016). Wang Thong District, where 10 sampling stations were established along irrigation canals sourced from the Wang Thong River, is characterized by fruit orchards and provides a contrasting agricultural land use setting for spatial comparison. The second comparison area, Mueang District (8 stations), was selected as a predominantly urban setting for comparison with the rice-dominated agricultural setting of Phrom Phiram (detailed GPS coordinates for all the sampling stations are provided in Supplementary Table S1). Accordingly, the sampling design was intended to characterize spatial contrasts among rice-dominated agricultural, fruit-orchard agricultural, and predominantly urban settings rather than to provide a probabilistic representation of all surface waters in Phitsanulok Province.

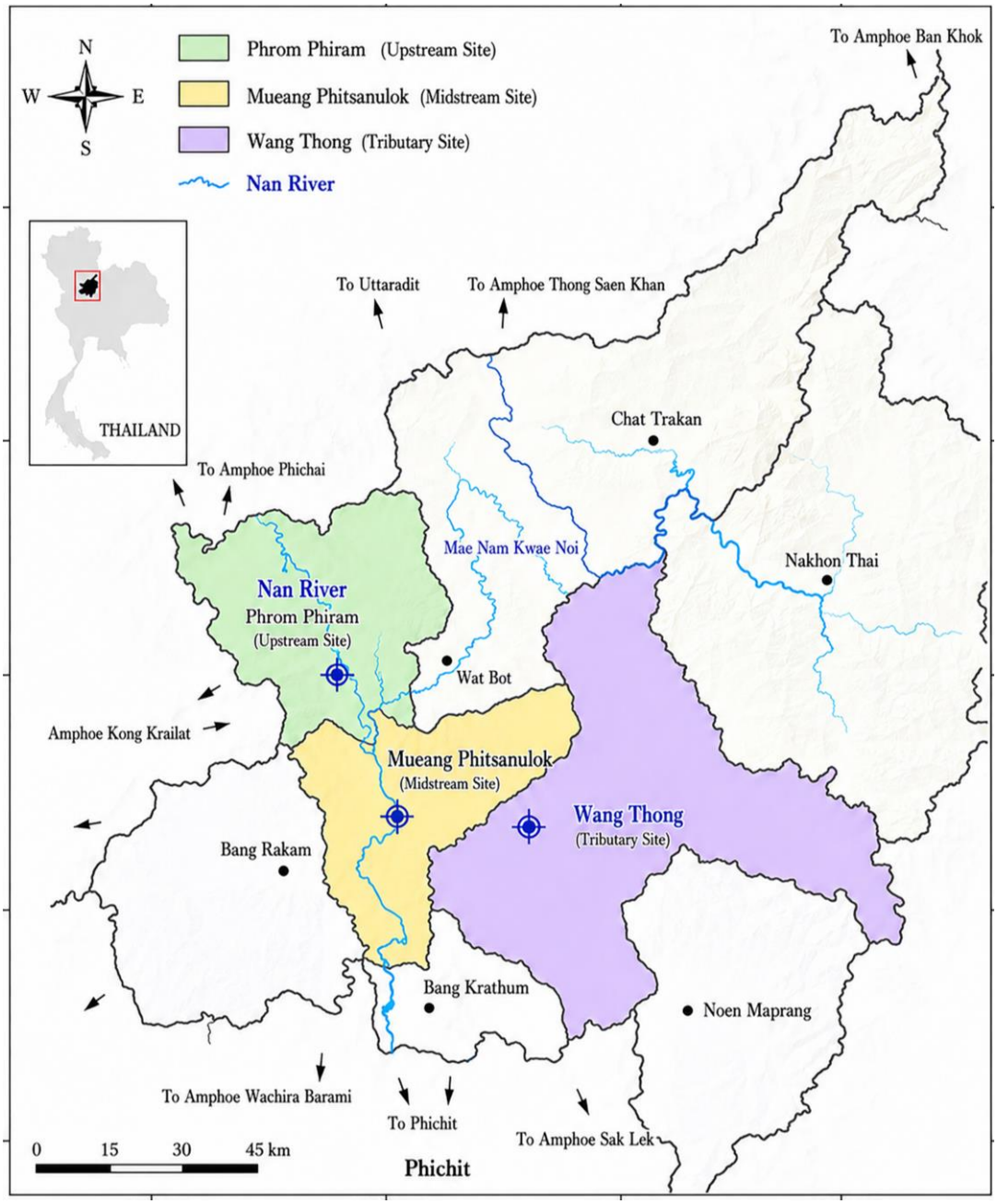


Figure 1 Map of the study area comprising three districts in Phitsanulok.

2) Sampling procedure and sample preservation

Water samples were collected using a grab sampling technique at mid-depth at each sampling station. Approximately 2 L of water was collected at each station using plastic containers, following standard surface-water sampling procedures (American Public Health Association, 1998; U.S. Environmental Protection Agency, 2013). All water quality parameters and paraquat concentrations were measured in triplicate. For phosphate analysis, one bottle was acidified with 65% nitric acid and stored at 4°C, while a second bottle, intended for general physicochemical analyses, was stored at 4°C until laboratory analysis.

3) Physicochemical analysis of water quality

The physical and chemical characteristics of the water samples were analyzed both in situ and in the laboratory following standard procedures (American Public Health Association, 1998). TDS and DO were measured in triplicate using a Multiparameter ProLab 2500 (SI Analytics, Weilheim, Germany) equipped with a conductivity probe and an FDO1100 IDS probe, respectively. The pH was measured in triplicate using an A162 IDS probe calibrated with standard buffer solutions at pH 4.0, 7.0, and 10.0. Measurements were automatically temperature-compensated and recorded at 25 ± 1°C. Turbidity was assessed with a HACH 2100P portable turbidity meter (Hach Company, Loveland, CO, USA) within the range of 0–800 NTU. For chemical analysis, phosphate concentrations were determined using the stannous chloride colorimetric method with a Biochrom Libra S70 spectrophotometer (Biochrom Ltd., Cambridge, UK) at 690 nm.

4) Paraquat determination

Paraquat concentrations in water samples were determined using a colorimetric test kit developed by the Department of Chemistry, Faculty of Science, Naresuan University (Phitsanulok, Thailand). This method is based on the chemical reduction of paraquat by sodium dithionite in an alkaline solution, which generates a characteristic blue-colored paraquat radical cation. Briefly, a 5 mL aliquot of each water sample was processed according to a previously described procedure (Sangsum and Saetear, 2022). The reaction was conducted at room temperature for 5 minutes. While the test kit allows for semiquantitative quantification, this study utilized spectrophotometric measurement at 395 nm using a Biochrom Libra S70 spectrophotometer (Biochrom Ltd., Cambridge, UK) to ensure quantitative accuracy. The working range of the method is 0.25 to 40 mg L^{-1}, with a limit of detection (LOD) of 0.25 mg L^{-1} (Sangsum and Saetear, 2022).

Calibration yielded a correlation coefficient (r) between 0.9984 and 1.000. Quality control was maintained by spiking samples (1 mg L^{-1}), with relative standard deviations (RSD) ranging from 1.14 ± 0.05% to 1.19 ± 0.04%. Method accuracy was evaluated using spiked samples at 1 mg L^{-1}, with recovery rates ranging from 75% to 120%, generally comparable with the recovery ranges reported in AOAC guidance for analytes at approximately the ppm concentration level (AOAC International, 2002).

5) Ecological risk assessment

To evaluate the potential ecological risks associated with paraquat residues in surface water, hazard quotients (HQs) were calculated using Eq. 1.

$$HQ = EEC / \text{Screening benchmark} \quad \text{(Eq. 1)}$$

where the environmental exposure concentration (EEC) represents the paraquat concentration (mg L^{-1}). For upper-bound screening, an estimated environmental concentration (EEC) was derived using Tukey's upper-fence approach (Q3 + 1.5 × IQR) (Tukey, 1977). In this study, this statistical threshold was used as an upper bound EEC for screening purposes. Thus, the upper EEC represents an upper-bound statistical screening estimate rather than a directly observed rainfall- or runoff-triggered peak concentration, given the absence of continuous event-based monitoring.

The ecological screening benchmark was set at 0.0012 mg L^{-1}, corresponding to the freshwater default guideline value (DGV) for paraquat at the 95% species protection level (Australian and New Zealand Governments, 2024). For the primary HQ calculations, nondetectable (ND) results were substituted with half the detection limit (0.13 mg L^{-1}), a commonly applied approach for censored environmental datasets when the true concentration below the detection limit is unknown. Since simple substitution may influence estimates derived from censored datasets, particularly when the proportion of nondetects is high (Hornung and Reed, 1990), the influence of this assumption was evaluated through a sensitivity analysis. The primary half-LOD scenario was therefore compared with an alternative zero-substitution scenario in which nondetects were assigned a value of zero (ND = 0). The resulting HQs were interpreted as screening-level estimates under analytical and censored-data uncertainty rather than as precise quantitative estimates of ecological risk. For screening-level interpretation, HQ > 1 was considered indicative of potential ecological concern, whereas HQ ≤ 1 indicated that the screening benchmark was not exceeded (Chele et al., 2023).

6) Household questionnaire survey

A total of 231 households were selected from Phrom Phiram Municipality (total population = 545 households) using a stratified random sampling technique, with the sample size determined using the

Yamane (1973) formula. Structured questionnaires were administered to household representatives to assess three primary areas: (1) sociodemographic factors (e.g., sex and age), (2) work characteristics (e.g., crop type and herbicide application history), and (3) general herbicide exposure practices and associated self-reported health symptoms. This comprehensive approach, rather than focusing exclusively on paraquat, was designed to reflect the concurrent postban use of alternative herbicides in the region. Specifically, this study focused on herbicides because they account for a substantial proportion of pesticide use and imports in Thailand (Kwonpongsagoon et al., 2021; Laohaudomchok et al., 2021). Participants were asked to identify their primary acute symptoms experienced within 24–48 hours and self-reported subacute symptoms occurring within 3 months following exposure. The respondents were instructed to select all primary or most prominent manifestations experienced, allowing for the identification of multiple concurrent symptoms. The questionnaire focused on prominent symptoms within defined post-exposure time frames to facilitate the reporting of acute and subacute health symptoms. Similar pesticide-related acute symptoms have been assessed among agricultural populations in previous studies (Desye et al., 2024; Sapbamrer et al., 2024). The instrument's validity and reliability were confirmed through the index of item-objective congruence (IOC), which ranged from 0.67 to 1.00, and a Cronbach's alpha coefficient ranging from 0.70 to 0.85. This study was conducted in accordance with ethical standards and approved by the Naresuan University Institutional Review Board (COA No. 356/2024).

7) Statistical analysis

Statistical analyses were performed using SPSS version 30.0. Descriptive statistics, including mean, median, minimum, and maximum values, were used to summarize paraquat concentrations and water quality parameters. For data points below the detection limit (ND), half the detection limit (0.13 mg L^{-1}) was used as the primary substitution approach for screening-level calculations. Since simple substitution can introduce bias in censored datasets, particularly when the proportion of nondetects is high (Helsel and Hirsch, 2002; Hornung and Reed, 1990), its influence was evaluated through a sensitivity analysis using an alternative zero-substitution scenario. The normality of the quantitative variables was assessed using the Shapiro–Wilk test. Given that the assumptions of normality were not met, the Kruskal–Wallis test was employed to compare paraquat concentrations and other water quality parameters across the three study areas (Phrom Phiram, Wang Thong, and Mueang Districts). Pairwise comparisons between these districts were subsequently conducted using Dunn's post hoc test, with a Bonferroni correction applied to adjust for multiple comparisons. Spearman's rank correlation was used to evaluate the relationships between paraquat levels and other physicochemical parameters. For all analyses, a p value < 0.05 was considered to indicate statistical significance.

Results and discussion

1) Physicochemical water quality and paraquat concentrations

The physicochemical profiles of surface water across the three study areas revealed near-neutral conditions, with pH values ranging from 7.42 to 7.57 at a controlled laboratory room temperature of 25 ± 1°C (Table 1). Phrom Phiram had the highest TDS (214.50 ± 25.70 mg L^{-1}) and turbidity (310.18 ± 127.25 NTU) among the three study areas. Phrom Phiram also had the highest phosphate concentration (0.47 ± 0.50 mg L^{-1}).

With respect to herbicide contamination, paraquat residues were most prevalent in Phrom Phiram (0.39 ± 0.06 mg L^{-1}), with a detection frequency of 75%. In contrast, lower concentrations and detection rates were detected in Wang Thong (0.18 ± 0.03 mg L^{-1}, 30%) and Mueang (0.16 ± 0.03 mg L^{-1}, 13%) (Supplementary Figure 1). The residual presence of paraquat in surface water in Phrom Phiram four years after the 2020 nationwide ban underscores a critical environmental challenge.

This observed residue likely reflects the historical intensity of herbicide use in Thai agriculture, where paraquat was among the most heavily imported and extensively applied herbicides prior to its nationwide ban (Kwonpongsagoon et al., 2021; Laohaudomchok et al., 2021). This environmental fate is consistent with previous studies highlighting the resistance of paraquat to microbial degradation and its prolonged persistence in soil, with reported half-lives of up to 6.6 years (Huang et al., 2019).

The environmental behavior of paraquat may provide a plausible context for the higher detection frequency observed in Phrom Phiram. Roberts et al. reported that the majority of paraquat that reaches soil under typical agricultural use conditions is strongly adsorbed to soil particles, leaving only a very low concentration in soil solution (Roberts et al., 2002). However, the paraquat present in soil solution remains susceptible to microbial degradation, and long-term field studies have shown that soil residues may eventually decline (Roberts et al., 2002).

Table 1 Water quality parameters and chemical and paraquat levels of water samples from 3 districts (Phrom Phiram, Wang Thong, and Mueang)

Parameters	District		
	Phrom Phiram	**Wang Thong**	**Mueang**
pH	7.47 ± 0.01	7.42 ± 0.03	7.57 ± 0.10 [a]
	7.47 (7.41–7.50)	7.46 (7.17–7.50)	7.70 (6.97–7.83) [b]
TDS (mg L^{-1})	214.50 ± 25.70	111.53 ± 5.24	172.96 ± 12.19
	219.17 (128.00–359.67)	110.00 (91.00–151.00)	166.00 (135.00–250.00)
Turbidity (NTU)	310.18 ± 127.25	75.08 ± 14.51	182.43 ± 106.75
	74.22 (40.70–770.00)	56.18 (31.00–169.00)	31.10 (12.30–803.00)
DO (mg L^{-1})	5.99 ± 0.20	5.67 ± 0.12	6.11 ± 0.18
	5.89 (5.17–6.94)	5.71 (5.02–6.36)	5.91 (5.48–6.92)
Phosphate (mg L^{-1})	0.47 ± 0.05	0.21 ± 0.20	0.20 ± 0.03
	0.53 (0.24–0.63)	0.21 (0.13–0.33)	0.16 (0.11–0.32)
Paraquat (mg L^{-1})	0.39 ± 0.06 [c]	0.18 ± 0.03	0.16 ± 0.03
	0.45 (0.13-0.67)	0.13 (0.13-0.29)	0.13 (0.13-0.33)

Remark: [a, b] The values are expressed as the mean ± SE and median (min–max), respectively.
[c] LOD values below the detection limit were substituted with LOD/2 (0.13 mg L^{-1}) for the primary screening-level calculations (Hornung and Reed, 1990).

Paraquat adsorption is also influenced by sediment properties and has been reported to be positively correlated with sediment cation exchange capacity (CEC) and clay content (Noicharoen et al., 2012). Environmental conditions can further modify this adsorption–desorption behavior. In river sediments, increased salinity has been reported to reduce paraquat adsorption and increase desorption, indicating that changes in water chemistry may facilitate the release of previously adsorbed residues (Noicharoen et al., 2012). Together with the prolonged environmental persistence reported in a previous study (Huang et al., 2019), these processes provide a plausible environmental context for the occurrence of paraquat residues in the surface waters of Phrom Phiram several years after regulatory prohibition. Nevertheless, the present study did not directly assess soil or sediment paraquat residues, adsorption–desorption processes, or the source and timing of contamination. Therefore, the higher detection frequency observed in Phrom Phiram cannot be attributed specifically to the remobilization of historically applied paraquat, and more recent or other unidentified inputs cannot be excluded.

2) The distribution of water quality parameters and paraquat levels across districts

The spatial patterns of water quality parameters and paraquat levels exhibited significant variability across the three study districts (Table 2). Significant differences in the TDS, phosphate, and paraquat concentrations were observed among the areas, whereas the pH, DO concentration, and turbidity (NTU) remained relatively consistent ($p > 0.05$). Notably, Phrom Phiram exhibited significantly higher levels of TDS and phosphate than Mueang and Wang Thong did ($p < 0.05$), indicating distinct water quality characteristics in this predominantly agricultural district.

Importantly, the distribution of paraquat followed a similar spatial pattern, with Phrom Phiram having significantly higher concentrations than Wang Thong ($p = 0.025$) and Mueang ($p = 0.010$). This heterogeneity suggests that paraquat residues were not uniformly distributed across the three study areas. Paraquat is strongly adsorbed to soil constituents, and its adsorption behavior is influenced by soil and sediment properties (Noicharoen et al., 2012; Roberts et al., 2002).

These findings align with those of previous studies indicating that soil texture and composition are important factors influencing paraquat adsorption (Flafel et al., 2024; Noicharoen et al., 2012). Strong adsorption to soil particles may contribute to the retention of paraquat residues in agricultural environments (Flafel et al., 2024; Roberts et al., 2002). Hydrological processes may subsequently facilitate the transport of particle-associated residues from agricultural soils to adjacent surface waters (Flafel et al., 2024). This mechanism provides a plausible environmental context for the occurrence of paraquat residues in Phrom Phiram; however, the specific source and transport pathways of the detected residues were not directly investigated in the present study. Our findings thus underscore the importance of continued postban environmental monitoring in areas with a history of intensive agricultural activity.

3) Correlations between water quality parameters and paraquat levels

The relationships between paraquat levels and selected water quality parameters were examined. Phosphate concentrations were significantly positively correlated with paraquat levels ($r = 0.426$, $p = 0.030$), indicating an association between the two variables.

Table 2 Comparison of physical, chemical, and paraquat levels in Phrom Phiram, Wang Thong, and Mueang districts

Parameters	Mean rank			Kruskal–Wallis H (df, *p* value)	Comparison of physical, chemical, and paraquat levels (*p value*)		
	Phrom Phiram	Wang Thong	Mueang		Phrom Phiram vs Wang Thong	Phrom Phiram vs Mueang	Mueang vs Wang Thong
pH	13.06	11.10	16.94	X^2 (2) = 2.63, (*p* = 0.269)	n.s. [a]	n.s.	n.s.
TDS (mg L^{-1})	19.69	5.80	16.94	X^2 (2) = 17.00, (*p* < 0.001)	< 0.001	n.s.	0.006
Turbidity (NTU)	17.94	8.81	13.70	X^2 (2) = 5.70, (*p* = 0.058)	n.s.	n.s.	n.s.
DO (mg L^{-1})	14.44	10.00	16.94	X^2 (2) = 3.83, (*p* = 0.147)	n.s.	n.s.	n.s.
Phosphate (mg L^{-1})	21.44	10.95	8.75	X^2 (2) = 12.87, (*p* =0.002)	0.011	0.003	n.s.
Paraquat (mg L^{-1})	19.75	11.35	9.94	X^2 (2) = 10.26, (*p* = 0.006)	0.025	0.01	n.s.

Remark: [a] n.s. (not significant) Dunn's post hoc test applied for pairwise comparisons.

This association may reflect shared agricultural influences rather than a direct interaction between phosphate and paraquat. Previous studies have shown that paraquat adsorption and desorption can be influenced by environmental conditions such as salinity, with increased salinity reducing adsorption and enhancing desorption from sediments (Noicharoen et al., 2012). Although parameters such as ionic strength and desorption kinetics were not directly measured in this study, these processes may influence the environmental behavior of paraquat. However, as correlation does not establish direct mechanistic causation, these observational trends should be interpreted with caution. Further studies under controlled conditions are needed to elucidate the mechanisms underlying the environmental fate and mobility of paraquat (Flafel et al., 2024; Noicharoen et al., 2012).

4) Ecological risk assessment

The ecological risk assessment indicated elevated screening-level HQs across all districts, with values exceeding the ecological screening benchmark (HQ > 1), as shown in Table 3. In the mean-exposure screening scenario, the HQ_{mean} values ranged from 125 to 325, with the highest value observed in Phrom Phiram. Under the upper-bound screening scenario, the HQ_{upper} values reached 742 in Phrom Phiram, followed by Wang Thong (442) and Mueang (108). These elevated HQs exceeded the ecological screening benchmark and indicate potential ecological concern under the upper bound EEC assumptions applied in this assessment. Paraquat residue may be particularly relevant to aquatic organisms, including primary producers, which are known to be sensitive to paraquat exposure (Donaher and Van den Hurk, 2023).

It is important to acknowledge that the HQ estimates were sensitive to the choice of substitution approach for nondetected values, particularly in districts with a high proportion of undetected observations. A sensitivity analysis comparing the primary half-LOD substitution (ND = 0.13 mg L^{-1}) with an alternative zero-substitution scenario (ND = 0) revealed that HQ estimates were influenced by the substitution approach used for nondetect observations. Under the zero-substitution scenario, the HQ_{upper} estimates differed from those obtained using the primary half-LOD approach. In Mueang District, HQupper decreased to zero because the interquartile range collapsed (IQR = 0). In contrast, Phrom Phiram and Wang Thong retained measurable variability (IQR = 0.38 and 0.29, respectively), resulting in HQ_{upper} values of 1.00 and 0.73, respectively. These findings indicate that the treatment of nondetect values influenced the distributional characteristics used to derive the upper EEC and subsequently affected HQ estimates. Therefore, HQ estimates should be interpreted as screening-level estimates that depend on the assumptions used for nondetected value treatment rather than as precise quantitative estimates of ecological risk. Furthermore, the use of Q3 + 1.5 × IQR to derive the upper EEC was used to derive an upper-bound screening estimate (Tukey, 1977). While this approach is useful for identifying upper-bound conditions, the lack of continuous event-based monitoring prevents direct inference of rainfall-triggered episodic exposure. The higher paraquat concentrations and detection frequency observed in Phrom Phiram may reflect historical paraquat use intensity in this rice-intensive landscape (Sornlorm et al., 2022) and support the need for continued temporal monitoring.

5) Agricultural practices and self-reported health symptoms among farmers in Phrom Phiram

The majority of the farmers in Phrom Phiram were male (71%), with an average age of approximately 49 years, as shown in Table 4. Rice cultivation was the most common single-crop practice among the participants (30%). Given that Phrom Phiram District exhibited the highest paraquat detection frequency (75%) and the highest screening-level HQs among the studied districts, this area was prioritized for the investigation of agricultural practices and farmer health. Analysis of the questionnaire data revealed a high prevalence of self-reported acute health symptoms; specifically, 69% (n = 160) of the participants experienced acute health symptoms within 24–48 hours following herbicide exposure, whereas no self-reported subacute symptoms occurring within up to 3 months were reported (0%). Among those reporting acute manifestations (n = 160, where respondents could report multiple symptoms), ocular and nasal irritation was the most prevalent (81%), followed by other nonspecific symptoms (19%), skin burns or irritation (14%), headache (8%), and vomiting (8%). While some of the reported symptoms are consistent with the known toxicological profile of paraquat (Sornlorm et al., 2022; Srisuma et al., 2023), these symptoms are nonspecific and should not be interpreted as evidence of paraquat-specific health effects. Given that no biomonitoring data (e.g., blood or urine analysis) or physician-confirmed diagnoses were collected in this study, a definitive causal link between environmental paraquat residues and farmer health symptoms cannot be rigorously established. These self-reported symptoms may also be associated with alternative herbicides or other environmental and occupational stressors encountered in agricultural settings; therefore, they should not be attributed specifically to paraquat exposure (Laohaudomchok et al., 2021; Sapbamrer et al., 2024).

Despite the high prevalence of self-reported acute health symptoms, 70% of individuals did not seek formal medical attention. This may suggest under-recognition or normalization of occupational health symptoms, although the reasons for not seeking medical care were not assessed in the present study. Similar acute pesticide-related symptoms have been reported among agricultural workers in previous studies (Desye et al., 2024; Sapbamrer et al., 2024), highlighting the broader occupational health relevance of these findings.

6) Limitations

Despite the detection of paraquat residues in surface water in this study, several critical limitations must be acknowledged. First, the colorimetric test kit, while practical for regional screening, has a limit of detection (LOD = 0.25 mg L^{-1}) (Sangsum and Saetear, 2022) that is substantially higher than the ecological screening benchmark (DGV = 0.0012 mg L^{-1}) (Australian and New Zealand Governments, 2024). This discrepancy indicates that concentrations below the LOD but above the benchmark could remain undetected, potentially leading to the underestimation of low-level contamination. Conversely, the substitution of nondetects with half the LOD (0.13 mg L^{-1}), which itself exceeds the ecological screening benchmark, may influence the calculated HQs, particularly in areas with a high proportion of undetected observations. Thus, the analytical limitation can introduce uncertainty in both directions: low-level residues may remain undetected, whereas substitution of nondetects may affect calculated HQs. Accordingly, the resulting HQs should be interpreted as screening-level estimates under analytical and censored-data uncertainty rather than precise quantitative estimates of ecological risk (Helsel and Hirsch, 2002).

Furthermore, environmental sampling was restricted to a single seasonal window (October–December), which may not fully capture the temporal variation associated with seasonal hydrological conditions. Additionally, as this study focused exclusively on surface water, it did not assess paraquat residues retained in soils or sediments, where paraquat can be strongly adsorbed and persist under environmental conditions (Huang et al., 2019; Noicharoen et al., 2012; Roberts et al., 2002).

Table 3 Hazard quotients (HQs) for paraquat by district using mean concentrations (chronic screening) and upper-bound EECs (Q_3 + 1.5×IQR; upper-bound screening scenario)

District	Mean concentration of paraquat (mg L^{-1})	Detection (%)	Upper EEC (mg L^{-1})	HQ_{mean}	HQ_{upper}
Phrom Phiram	0.39 ± 0.06[a]	75	0.89	325	742
Wang Thong	0.18 ± 0.03	30	0.53	150	442
Mueang	0.16 ± 0.03	13	0.13	125	108

Remark: [a] The values are expressed as the mean ± SE.

Table 4 Characteristics of the study subjects and herbicide use and related self-reported health symptoms

Parameters	n (%)
Sociodemographic factors	
Gender	
Male	163 (71%)[a]
Female	68 (29%)
Age (years)	48.86 ± 0.58[b]
	50.00(25.00-59.00)[c]
Type of crop cultivation	
- Rice	69 (30%)
- Chili Peppers	45 (19%)
- Corn	15 (6%)
- Culinary herbs and garden Vegetables	10 (4%)
- Other (e.g., multiple crops)	92 (40%)
Years of herbicide use	6.05 ± 0.17
	5.00(1.00-15.00)
Herbicide application patterns	
Continuous (Year-round)	66 (29%)
Targeted (Pre and postharvest)	165 (71%)
Self-reported acute health symptoms (n = 231)	
Presence of acute symptoms	160 (69%)
Absence of acute symptoms	71 (31%)
Subacute symptoms (n = 231)	
Presence of subacute symptoms	0 (0%)
Reported acute symptoms (n = 160)	
- Eye and nasal irritation	130 (81%)[d]
- Skin burns/irritation	22 (14%)
- Headache	13 (8%)
- Vomiting	13 (8%)
- Other	30 (19%)
Healthcare seeking behavior (n = 231)	
- Seek medical attention	69 (30%)
- Don’t seek medical attention	162 (70%)

Remark: [a] The values are expressed as the number of subjects (percentage).
[b, c] The values are expressed as the mean ± SE and median (min–max), respectively.
[d] Participants could experience/report multiple symptoms; thus, the sum of the percentages exceeded 100%.

Consequently, future studies should employ high-sensitivity analytical techniques, such as LC–MS/MS, to more accurately quantify low-level paraquat residues below the detection capability of the current screening method. Multiseason monitoring covering wet, dry, and agricultural irrigation periods is also recommended to characterize the temporal variability in paraquat occurrence and ecological risk.

With respect to health assessments, reliance on self-reported symptoms introduces potential recall bias (Sapbamrer et al., 2024). The lack of biomonitoring data precludes the establishment of a direct causal link between environmental paraquat concentrations and the reported incidence of acute symptoms. Other environmental and occupational stressors were not measured and may have contributed to the reported symptoms (Laohaudomchok et al., 2021; Sapbamrer et al., 2024).

Conclusions

This study demonstrated that paraquat residues remained detectable in the surface waters of Phitsanulok Province several years after Thailand’s 2020 national ban, with the highest detection frequency observed in the rice-dominated agricultural setting of Phrom Phiram. The moderate positive correlation between phosphate and paraquat indicates an association between the two variables; however, the underlying environmental processes responsible for this relationship cannot be determined from the present data.

Ecological risk screening indicates potential concerns for aquatic ecosystems across the studied

districts, with hazard quotients exceeding the screening threshold (HQ > 1) under the upper-bound EEC approach applied in this assessment. Furthermore, while a high prevalence of acute health symptoms was reported among surveyed farmers, these symptoms were self-reported and nonspecific and should not be interpreted as evidence of paraquat-specific health effects. The low rate of healthcare seeking despite the reported symptoms may suggest underrecognition or normalization of occupational health symptoms and warrants further investigation. Collectively, these findings indicate that paraquat residues can remain detectable in agricultural surface waters several years after regulatory prohibition, although the specific source and timing of the detected residues could not be determined. Beyond screening-level assessments, future studies employing LC–MS–MS or other high-sensitivity analytical methods together with multiseason environmental monitoring will be important for better characterizing low-level paraquat residues, temporal variability, and associated ecological concerns in postban agricultural settings. Further investigations of occupational health risks and healthcare-seeking behavior are needed to better understand potential health concerns in intensive agricultural settings.

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Data availability statement

Information and data used in the study will be disclosed upon request.

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Conflicts of interest

The authors declare that there are no conflicts of interest in competing financial or personal relationships that could have appeared to influence the work reported in this work.

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