

Screening analysis of organic micro-pollutants in groundwater in Northern Vietnam: Occurrence, sources and risk assessment

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Abstract

Groundwater, a primary drinking water source in Vietnam, is threatened by increasing contamination from organic micropollutants (OMPs). We applied an automated identification and quantification system (AIQS) with gas chromatography- (GC-MS) and liquid chromatography-time of flight (LC-QTOFMS) mass spectrometry databases to screen ~ 1500 OMPs in groundwater in Vietnam. Due to sample transport restrictions, an international sample transfer method ("travel analysis") using solid-phase extraction cartridges was applied to adsorb and transport analytes in samples from Vietnam to Japan. Designated quality control tests confirmed the stability of multiple analytes during travel analysis. This method was applied to investigate OMPs occurrence in 19 well water samples collected from urban, suburban, agricultural, and industrial areas in northern Vietnam. OMPs were observed in all samples, with 60 OMPs being detected. OMPs (~ 40) were identified for the first time in the Vietnamese groundwater samples. Pharmaceuticals, personal care products (PPCPs), plasticizers, and fire retardants were frequently detected. Higher levels of OMPs contamination were observed in urban and agricultural areas than those in the other areas. Specifically, PPCPs and plasticizers were more frequently found in agricultural wells, whereas fire retardants, pesticides, and household-use chemicals were more prevalent in urban sites. In addition to infiltration from surface sources, another possible route for OMP to reach groundwater is suggested via leaky boreholes. The coexistence of persistent and short-lived OMPs revealed that Vietnamese aquifers likely face both ongoing and legacy contamination. No significant health risks were observed based on our results.

1. Introduction

Freshwater is essential for human life, yet accessible drinking water constitutes only a small proportion of the total water resources of the Earth and is mostly stored in aquifers. Groundwater is characterized by a high organoleptic quality and supplies nearly half of the global population (Siebert et al. 2010). Aquifers are primarily recharged through seepage from surface sources such as rain, rivers, and lakes. Consequently, aquifers are vulnerable to surface pollution, particularly from organic micropollutants (OMPs) (Mackay and Roberts 1985). Earlier studies identified several OMPs and their metabolites in aquifers across many regions globally, including European (EU) countries (Bunting et al. 2021), the United States (US) (Barnes et al. 2008), and China (Kong et al. 2016; Li et al. 2020). Many OMPs, such as pesticides, polycyclic aromatic hydrocarbons (PAHs), pharmaceuticals, personal care products (PPCPs), fire retardants (FRs), plasticizers, perfluoroalkyl substances, and other emerging organic pollutants, were identified or are suspected to cause adverse human health effects, including endocrine disruption and carcinogenicity (Bunting et al. 2021).

Vietnam is a developing country in Southeast Asia where agriculture accounts for a significant proportion of the economy. Over the last decade, industrialization and urbanization in Vietnam have increased sharply, increasing the burden on the environment, especially in aquatic environments. Earlier studies reported increasing contamination by diverse OMPs in surface water and sediments, particularly in urban, industrial, and agricultural zones (Chau et al. 2018; Duong et al. 2015b; Truong et al. 2023).

The average estimated water demand in Vietnam was more than a million m³ per day (in 2010) (Moglia et al. 2012), of which groundwater exploitation was approximately half (Duong et al. 2015a). Three main types of groundwater exploitation exist in Vietnam: major wells for city water supply treatment plants, medium private wells for business and factory operations, and small private wells for household use (Duong et al. 2015a). To the best of our knowledge, few studies were conducted on the occurrence of organic contaminants in groundwater in Vietnam. These investigations focused primarily on urban areas and used a screening method with gas chromatography-mass spectrometry (GC-MS) (Duong et al. 2015a) or targeted analysis methods with liquid chromatography (Duong et al. 2015b; Lam et al. 2017). These studies detected only a subset of contaminants, with no updated surveys over the past decade. Information on OMPs in vulnerable areas, such as industrial and agricultural zones, remains unknown. Particularly, the groundwater usage rates in these zones in Vietnam are high. Additionally, in recent years, the development of advanced analysis methods, such as the Automated Identification and Quantification System (AIQS) with liquid chromatography-time of flight mass spectrometry (LC-QTOFMS), has allowed the simultaneous detection of ~ 500 OMPs (Kadokami et al. 2023), providing opportunities for comprehensive assessment. Furthermore, groundwater contamination profiles change over time with changes in human activities (Lindsey et al. 2021). Recent industrialization and urbanization in Vietnam may have altered patterns of chemical use, thereby shifting groundwater contamination profiles. Therefore, research on the occurrence and behavior of OMPs in groundwater from different areas in Vietnam using the latest analytical methods is urgently needed to understand the current contamination status, land-use influences, and potential human health risks.

The AIQS with Database (AIQS-DB) method is reliable, quick, and cost-effective for identifying and semi-quantifying various organic substances in different environmental samples (Kadokami et al. 2005; Kong et al. 2016). This method was successfully applied for the analysis of hundreds of OMPs in surface and groundwater samples (Chau et al. 2018; Duong et al. 2015a; Kong et al. 2016). To date, we have continued to develop this method, and the total number of registered OMPs in the database has expanded to ~ 1500 compounds.

To understand the presence of OMPs in groundwater and their potential risks to human health in different areas of northern Vietnam, AIQS with GC-MS and LC-QTOFMS databases was used to screen ~ 1500 OMPs in groundwater samples. To use this method, the analysis must be performed in a laboratory in Japan. However, the water samples collected in Vietnam cannot be transported to Japan. Therefore, it is necessary to have an international sample transfer ("travel analysis") process to overcome this issue. Some earlier studies attempted to use solid-phase extraction (SPE) for the transport of analytes in water samples (Carlson et al. 2013; S`liwka-Kaszyńska et al. 2003). However, only a few reports evaluated the stability of a limited number of compounds during storage on SPE cartridges (Carlson et al. 2013; S`liwka-Kaszyńska et al. 2003). Therefore, insufficient evidence exists to confirm the effectiveness of this approach in preserving a broad range of organic compounds with diverse physicochemical properties during transportation. Here, we attempted to solve this travel analysis issue using SPE cartridges. The analytical species were transported from Vietnam to Japan, adsorbed onto SPE cartridges, and analyzed in Japan. To achieve this, it was necessary to verify the stability of the analytical species during travel analysis.

Here, we aim to (1) verify the stability of analytes during the travel analysis process, (2) determine the pollution status of various OMPs in groundwater in northern Vietnam, (3) identify the suspected origins of the detected OMPs, and (4) estimate the potential health effects posed by OMPs to groundwater consumers. Our results will provide valuable information not only for Vietnam but also for other Southeast Asian countries using groundwater for drinking, and will provide important data for taking measures to conserve groundwater.

2. Materials and methods

2.1. Chemicals and materials

The solvents used were pesticide residues or liquid chromatography grade, provided by Kanto Chemical Co. Inc., Japan; FUJIFILM Wako Pure Chemical Co., Japan; or Fisher Chemical, UK. The OASIS HLB Plus LP extraction cartridges (HLB) and Sep-Pak Plus AC-2 cartridges (AC-2) were purchased from Waters Co. (Ireland). The 60 mm glass microfiber filters (GF/A) were supplied by Cytiva, China. Sodium sulfate anhydrous of pesticide residue quality were provided by Kanto Chemical Co. Inc., Japan. Buffer solution of $\text{NaH}_2\text{PO}_4\text{-Na}_2\text{HPO}_4$ (1 mol L^{-1} , pH 7.0) were also prepared. The nitrogen gas used was 99.999% pure. Millipore syringe filters (Millex-LG, $0.2 \mu\text{m}$, 4 mm; PTFE, Hydrophilic) were purchased from Merck Millipore. All glassware were rinsed with the appropriate solvents at least twice before use.

2.2. Sample collection

The study area is located in the Red River delta in northern Vietnam and encompasses urban, suburban, industrial, and agricultural areas. Hanoi is the largest and most crowded city in Vietnam. Urban and suburban Hanoi was chosen to represent the respective areas. The Yen My and My Hao districts in Hung Yen province were selected as industrial zones. In this zone, four large industrial parks exist within the study site discharging wastewater into rivers and canals. The main production types in industrial parks include food production; textiles, leather, and related products; wood processing; pharmaceuticals and medicinal materials; manufacturing of products from plastics and metals; and vehicle assembly. Thanh Mien district in Hai Phong province and Quynh Phu district in Hung Yen province were chosen as agricultural sites for this study. In these areas, more than half of the land is used for the cultivation of rice and vegetables, whereas the remaining areas are dedicated to residential purposes, aquaculture, animal husbandry, and small workshops.

The Red River delta is a large alluvial plain with mostly similar hydrogeographic characteristics, and the topmost aquifer is Holocene unconfined, occupying 88% of the delta (Bui et al. 2011). Therefore, all study sites were selected to have similar hydrogeographical characteristics, with most soils at the sites being clay slurry, sandy dust, and gray sand with high permeability (Bui et al. 2011; NAWAPI 2015).

Nineteen groundwater samples were collected from 19 domestic wells at the study sites: urban ($n = 5$), suburban ($n = 3$), industrial ($n = 5$), and agricultural ($n = 6$) (Fig. 1, Table S1). Given the similar hydrogeological characteristics across the study areas (Bui et al. 2011), sampling points near the main streams, central locations, or notable land use sites were selected.

The bottles for groundwater sampling were pre-washed with solvents before use: for GC analysis, 1 L dark glass bottles pre-washed with acetone, hexane, and purified water was used; for LC analysis, 500 mL polyethylene terephthalate (PET) bottles pre-washed with methanol and purified water were used. To ensure the representativeness of the samples in all wells, groundwater was pumped at least 5 min prior to collection, and the sampling bottles were soaked at least five times with water samples before sampling. For each site, a total of 1.5 L of raw groundwater was collected in a 1 L dark glass bottle and a 500 mL PET bottle. The samples were then labeled, stored in cooler boxes with ice packs, and transported to the laboratory at the Institute of Science and Technology for Energy and Environment (ISTEE), Vietnam, on the same day. Upon arrival, samples were stored at $4^\circ\text{C} \pm 2^\circ\text{C}$ for maximum 2 days before the further steps.

2.3. Sample extraction and analysis

2.3.1. Sample preparation in Vietnam

Sample filtration and solid-phase adsorption were conducted in a laboratory in Vietnam. The details are described below.

For GC-MS analysis of ~ 969 OMPs (Table S2) in the groundwater samples, the method of MoE (2023) was used with some modifications for OMPs extraction. Briefly, the HLB and AC-2 cartridges were preconditioned with 10 mL of dichloromethane (DCM), 10 mL of acetone, and 20 mL of ultrapure water. The groundwater sample (500 mL) was filtered through a GF/A filter after adjusting the pH to 7 using a buffer solution ($\text{Na}_2\text{HPO}_4\text{-NaH}_2\text{PO}_4$). The filtrate was applied to HLB and AC-2 cartridges at a flow rate of 3 mL min^{-1} using a vacuum manifold (Supelco, Inc.). Then, the cartridges were fully dried under nitrogen stream at pressure of 0.3–0.4 MPa for 30 min.

The method described by Kadokami et al. (2023) was used for the LC-QTOF-MS analysis of ~ 540 OMPs (Table S3) in groundwater samples. Briefly, the HLB and AC-2 cartridges were preconditioned with 5 mL DCM, 5 mL methanol, and 10 mL ultrapure water. The pH of the sample (200 mL) was adjusted to 7 by adding phosphate buffer before filtration through GF/A. The filtrate was then passed through the HLB and AC-2 cartridges at a flow rate of 3 mL min^{-1} using a vacuum manifold. Thereafter, the cartridges were fully dried under nitrogen stream at pressure of 0.3–0.4 MPa for 30 min.

2.3.2 Sample storage and transfer

The SPE cartridges were capped and wrapped in aluminum foil. The caps were pre-cleaned with acetone and ultrasonicated for 10 min. The filtered GF/A were wrapped in an acetone-washed aluminum foil. Then cartridges and GF/As were stored at $4^\circ\text{C} \pm 2^\circ\text{C}$ in a fridge for maximum 10 days in Vietnam. The

cartridges and GF/As were then transferred to tightly sealed cooler boxes and kept cool with ice packs during transport for a maximum of 10 h to the laboratory at the University of Kitakyushu (UKK), Japan. Upon arrival, the samples were stored at $4^{\circ}\text{C} \pm 2^{\circ}\text{C}$ for maximum 10 days before further steps.

2.3.3. Sample elution and instrument analysis

Before elution, all samples, including cartridges and GF/As, were thawed in room temperature for 15 min, then cartridges were dried under nitrogen stream at pressure of 0.3–0.4 MPa for 5 min.

For GC-MS analysis, GF/A was extracted twice by ultrasonic extraction using 3 mL of acetone and 3 mL of DCM. The extract was concentrated to 1 mL under a gentle nitrogen stream. Then, 1 mL of hexane and 4 mL of ultrapure water were added, soaked well, and the upper layer of solution was taken. The same procedure was repeated once by adding 1 mL hexane, and the extracts were combined and dehydrated using anhydrous sodium sulfate. The cartridges were eluted separately: HLB cartridges with 2 mL acetone and 3 mL DCM, and AC-2 cartridges with 3 mL acetone. The eluted solution from the cartridges and the GF/A extract were then combined and concentrated to 1 mL under a gentle nitrogen stream. After the solvent was changed to hexane, the solution was concentrated to 1 mL under gentle nitrogen stream, and added 100 μL of internal standard (custom internal standard mix, Restek, Japan, Pro. no. 578511) (Table S4). Instrumental analysis was performed using GC-MS-Scan (GCMS-TQ8040, Shimadzu, Japan), following the procedures and conditions described by the MoE (2023) (Table S5).

For LC-QTOF-MS analysis, the suspended solids on GF/A were extracted twice using ultrasonic extraction with 3 mL of methanol. The dried cartridges were eluted simultaneously (backflush) with 5 mL methanol and 3 mL DCM. The eluted solution from cartridges and extract of GF/A were combined and was then concentrated to 400 μL . Then, 50 μL of internal standards (AIQS-LC internal standard mix, HPC, Japan, Pro. no.: 99056217) (Table S4) was added, and the extract was reconstituted to 500 μL with methanol. Finally, the mixture was filtered through a 0.2 μm syringe filter prior to LC-QTOF-MS (X500R, SCIEX) analysis, using instrument parameters described by Kadokami et al. (2023) (Table S6).

2.4. Method validation and quality control

To validate the stability of the analytes adsorbed in SPE cartridges during the storage and transfer processes from Vietnam to Japan, quality control was performed through recovery tests for both AIQS with GC-MS and LC-QTOFMS analyses, which were conducted in the laboratory at the University of Kitakyushu. However, because it is not feasible to examine large numbers of the ~ 1500 compounds registered in the AIQS database, two standard mixtures containing multiple pesticides with a wide range of log P values were employed for recovery testing. Details of the standard chemicals are provided in Table S7. Pesticides are well-characterized, analytically diverse, and provide suitable standard chemicals to evaluate the method performance for a broad spectrum of organic chemicals.

For GC analysis, a mixture of 66 pesticide standards - Kanto Chemical Co., with log P range of 0.51–7.0 (Table S7), was used. Three standard samples were prepared by adding 0.5 mL buffer solution and 100 μL of standard mixture in 500 mL of ultrapure water.

For LC analysis, a mixture of 204 pesticide standards - LC Multiresidue Pesticide Standards Kit - Restek Corp., with log P range of -1.67–7.16 (Table S7), was used. Three standard samples were prepared by adding 0.5 mL buffer solution and 50 μL of standard mixture in 200 mL of ultrapure water.

The samples were processed following the normal adsorption procedure on SPE cartridges. The cartridges were stored under conditions simulating the actual storage and transport processes from Vietnam to Japan. Specifically, cartridges were first stored in a refrigerator at $4 \pm 2^{\circ}\text{C}$ for 10 days as designated storage condition in Vietnam. To mimic the transport process, the samples were placed in a cooler box with ice-gel packs for 10 h. Upon arrival for simulation in Japan, the cartridges were returned to refrigerated storage at $4 \pm 2^{\circ}\text{C}$ for another 10 days. Finally, the cartridges were eluted and analyzed following a normal procedure.

Blank samples were treated using the same procedure as that for the environmental samples, but with purified water instead of groundwater. During data reporting, the blank concentration (Table S8) was subtracted from the detected value of the samples; however, the concentration of detected compounds less than twice that in the blank was considered not detected (n.d.). The method detection limits of the individual chemicals for AIQS with GC-MS and LC-QTOFMS were 2–400 ng L^{-1} (Table S2) and 0.004–100 ng L^{-1} (Table S3), respectively.

2.5. Statistical analysis

Microsoft Excel version 2021, Microsoft, Redmond USA, WA., and IBM SPSS Statistics ver. 26, IBM Japan, was used for the statistical data analysis. Principal component analysis (PCA) was conducted using SPSS, where only chemicals detected with a detection frequency (DF) higher than 20% were calculated, and values below the method detection limit ($< \text{MDL}$) were set to zero. Statistical significance was set at $P < 0.05$.

2.6. Health risk assessment

The potential health risk, expressed as the risk quotient (RQ) for individual pollutants, was estimated following the guidelines of the US Environmental Protection Agency (USEPA 1989):

$$\text{RQ}_i = \text{CDI}_i / \text{RfD}_i \quad (1)$$

where, RfD_i is the chronic reference dose for an individual chemical ($\text{mg kg}^{-1} \text{ day}^{-1}$). The RfD values were collected from the US EPA Integrated Risk Information System (IRIS 2025) and US Risk Assessment Information System (RAIS 2024). If the RfD was not available, the calculated RfD based on the NOAEL (no observed adverse effect level) or LOAEL (lowest observed adverse effect level) (Li et al. 2020) was used instead. When the NOAEL and LOAEL are unavailable, the RfD values predicted by the quantitative structure–activity relationship (QSAR) models in the conditional toxicity value (CTV) software

(Wignall et al. 2018) can be employed. CDI_i is the chronic daily intake of a chemical ($\text{mg kg}^{-1} \text{ day}^{-1}$) and was calculated using the following equation (USEPA 1989):

$$CDI_i = (C_i \times IR \times EF \times ED) / (BW \times AT \times 10^6) \quad (2)$$

where, C_i is the measured concentration of a chemical in the environmental sample (ng L^{-1}). IR is the ingestion rate, set as 2 L day^{-1} . EF is the exposure frequency, which was set to $365 \text{ days year}^{-1}$. ED is the exposure duration, which was set to an average of 70 years. BW is the body weight, with an average weight of Vietnamese adults is 60 kg (Anh et al. 2019). AT is the average lifetime ($ED \times 365 \text{ days}$). From the estimated RQs, if the value is equal to or higher than 1, potential effects may exist on consumer health; else, no significant risk is observed (USEPA 1989).

3. Results and discussion

3.1. Validation of analytical method

For the AIQS with GC-MS analysis, the results of the recovery tests were evaluated according to a provisional manual issued by the Ministry of the Environment, Japan (MoE, 2023). Based on the provisional manual, target recovery and relative standard deviation (RSD) were set as 50–200% and $\leq 20\%$, respectively (MoE, 2023). Of the 66 standard substances, 65 (98%) met the criteria, with recoveries ranging from 61–115% and an average of 90%. The RSDs of these chemicals were below 20%, with an average of 7% (Table S7).

For AIQS with LC-QTOFMS, the results of the recovery tests were assessed by comparison with the Guidelines for Validation of Analytical Methods, Ministry of Agriculture, Forestry and Fisheries, Japan (MAFF 2023). Based on the guideline, target recovery and RSD were set as 40–120% and $\leq 20\%$, respectively (MAFF 2023). Among the 204 standard substances, 187 substances (92%) achieved the target recovery rate, with recoveries varying from 40–114%, with an average of 84%, and RSDs of 1–20%, with an average RSD of 6% (Table S7). Five other chemicals were not detected, and 12 showed recovery rates of 22–39%.

A small fraction of the analytes fell outside the acceptable range. Some highly hydrophobic compounds, such as eprinomectin and doramectin, exhibited low or inconsistent recoveries, probably because of their strong sorption onto glassware or filters and reduced solubility during sample preparation. Conversely, unstable or highly polar compounds such as trichlorfon and omethoate showed poor recoveries or high RSDs. Trichlorfon is an unstable insecticide easily decomposing under analytical conditions (Wang et al. 2010), and omethoate is a highly polar compound difficult to extract from water samples using SPE cartridges (Ingelse et al. 2001).

The RSD values in both tests were consistently low, with RSDs of $< 10\%$ for most of the standard substances. This low variability confirmed the reliability and reproducibility of the methods, ensuring that the quantitative differences observed in the environmental samples were meaningful. This robustness is essential for environmental monitoring in Vietnam, where complex mixtures of hydrophobic and hydrophilic compounds coexist. The minor limitations identified here are unlikely to affect the overall screening capacity.

The recovery tests for AIQS with GC-MS and LC-QTOFMS showed reliable results, confirming that the travel analysis method using AIQS with GC-MS and LC-QTOFMS is sufficient for the broad-spectrum screening of OMPs in water samples collected in Vietnam and measured in Japan. The high coverage of compound classes makes this method convenient for assessing contamination dynamics in aquatic environments in developing countries, such as Vietnam, where organic substance usage is diverse and regulations remain limited.

3.2. Overview of pollution status of OMPs in groundwater

The analysis of 19 groundwater samples from northern Vietnam identified 60 OMPs, including 19 pesticides, 7 pharmaceuticals and personal care products (PPCPs), 4 fire retardants (FRs), 5 plasticizers, 3 sterols, and 22 other chemicals (Table 1, Fig. 1). Of these, 26 OMPs were identified by AIQS with LC-QTOFMS and 34 were identified by GC-MS, with ~ 40 compounds reported for the first time in Vietnamese groundwater samples. The number of detected OMPs per sample ranged from 4–23, with a median of 9. The total concentrations of detected OMPs (ΣOMPs) varied between 201 and $11,200 \text{ ng L}^{-1}$, with a median of $1,370 \text{ ng L}^{-1}$. The maximum ΣOMPs was observed at G15, which was lower than those previously reported in urban Vietnam in 2014 ($16,000 \text{ ng L}^{-1}$) (Duong et al. 2015a) and China ($75,000 \text{ ng L}^{-1}$) (Kong et al. 2016). Higher OMPs contamination was observed in urban and agricultural areas, with 39 and 38 detections and medians of $1,770 \text{ ng L}^{-1}$ and $1,850 \text{ ng L}^{-1}$, respectively, compared with those of suburban (21 detections; $1,370 \text{ ng L}^{-1}$) and industrial areas (16 detections; 616 ng L^{-1}). This spatial pattern may suggest the substantial influence of domestic activities and agricultural production on groundwater pollution.

The detection frequencies (DFs) of the 60 OMPs ranged from 5–100%, with individual concentrations varying from $< \text{MDL}$ to $10,200 \text{ ng L}^{-1}$. However, 94% of the detected values were less than $0.1 \mu\text{g L}^{-1}$. PPCPs and plasticizer groups were present in more than 95% of the wells, followed by FRs (68%), sterols (47%), and pesticides (47%). The ubiquitous spread of these chemicals implies that they are commonly used in daily life and production activities in the study area.

Variations in the contamination profiles may reflect overlapping influences. Industrial zones were affected not only by manufacturing, but also by residential and small-scale farming, while agricultural areas were influenced by both farming practices and domestic discharge. Such overlapping pressures may explain the diverse mixtures of the detected OMPs. The presence of a wide spectrum of compounds emphasizes the vulnerability of the aquifers to pollution from various anthropogenic sources. Nevertheless, site-specific differences in land use, infrastructure, and wastewater management resulted in spatial variability in both the types and levels of OMP contamination, as discussed in subsequent sections.

Table 1

Summary of concentrations (ng L⁻¹) of organic micro-pollutants (OMPs) in groundwater in Vietnam and the

No.	Detector	Origin	Group	Name	Log P	DF %	Surburban			Urban					
							G1	G2	G3	G4	G5	G6	G7	G8	
1	GC-MS	Pesticide	Insecticide	3-Hydroxycarbofuran	1.2	16	1690	-*	-	-	-	-	-	1970	
2	LC-QTOFMS			Benzoximate	4.7	5	-	-	-	-	-	1.18	-	-	
3	LC-QTOFMS			Emamectin-benzoate b1a	5	5	3.14	-	-	-	-	-	-	-	
4	LC-QTOFMS			Etoxazole	5.59	5	-	-	-	-	-	1.97	-	-	
5	LC-QTOFMS			Furathiocarb	4.7	5	-	-	-	-	-	3.27	-	-	
6	LC-QTOFMS			Triflumuron	5.6	5	-	-	-	-	-	-	-	-	
7	LC-QTOFMS		Fungicide	Carbendazim	1.52	5	-	-	-	0.58	-	-	-	-	
8	LC-QTOFMS			Cyprodinil	3.59	11	-	-	-	<MDL	0.8	2.7	-	-	
9	LC-QTOFMS			Fenazaquin	5.51	5	<MDL	<MDL	-	<MDL	<MDL	<MDL	-	<MDL	
10	LC-QTOFMS			Fenpropimorph	5.2	5	-	-	-	-	-	6.22	-	-	
11	LC-QTOFMS			Pencycuron	4.68	5	-	-	-	-	-	2.34	-	-	
12	LC-QTOFMS			Picoxystrobin	3.6	5	-	-	-	-	-	0.92	-	-	
13	LC-QTOFMS		Herbicide	Trifloxystrobin	4.5	5	-	-	-	-	-	1.48	-	-	
14	LC-QTOFMS			Atrazine	2.61	5	-	-	-	49.8	-	-	-	-	
15	LC-QTOFMS			Atrazine-desethyl	1.7	5	-	-	-	7.56	-	-	-	-	
16	LC-QTOFMS			Diuron	2.68	5	-	-	-	-	-	-	-	-	
17	LC-QTOFMS			Metolachlor	2.9	5	-	-	-	-	-	-	2.90	-	
18	GC-MS			Sterols	Other	Oxabetrinil	2.6	5	18.5	-	-	-	-	-	-
19	GC-MS	Ethychlozate	0.1			5	-	-	-	-	-	-	-	-	
20	GC-MS	Stigmasterol	8.8			26	-	-	-	-	-	-	514	-	
21	GC-MS	Ergosterol	8			5	-	-	-	-	-	-	-	-	
22	GC-MS	Cholesterol	8.3			47	<MDL	-	53.9	-	-	66.7	97.5	<MDL	
23	LC-QTOFMS	PPCPs	Theophylline			0.1	11	-	-	-	-	-	-	-	-
24	GC-MS			2-Ethyl-1-hexanol	2.9	89	365	82	159	157	56.3	121	-	103	
25	GC-MS			2-phenoxy-Ethanol	2.2	37	-	-	-	-	-	-	-	83.7	
26	GC-MS			Diethyltoluamide	2.4	58	<MDL	<MDL	5.83	<MDL	<MDL	7.75	8.55	4.39	
27	GC-MS			Benzophenone	3.2	11	5.35	-	-	-	-	-	-	-	
28	LC-QTOFMS			Ethylhexyl methoxycinnamate	5.7	11	-	-	-	-	-	-	-	-	

No.	Detector	Origin	Group	Name	Log P	DF %	Surburban		Urban					
							G1	G2	G3	G4	G5	G6	G7	G8
29	LC-QTOFMS			Octocrylene	6.9	26	-	-	72.0	-	-	-	11	-
30	GC-MS	Fire retardants		Tris(1-chloro-2-propyl)phosphate	2.5	37	-	-	137	-	138	-	235	62.9
31	LC-QTOFMS			Tri-n-butyl phosphate	4	26	24.0	-	19.8	-	25.8	-	-	26.4
32	LC-QTOFMS			Trisisobutyl phosphate	4.1	26	51.6	-	43.3	-	55.2	-	-	56.4
33	GC-MS			Triphenyl phosphate	4.6	47	67.7	27.7	-	-	-	-	98.8	-
34	GC-MS	Plasticizer	Phthalate	Butyl benzyl phthalate	4.7	32	-	-	-	-	15.4	-	53.5	-
35	GC-MS			Di-iso-butyl phthalate	4.1	26	-	-	-	-	-	-	325	-
36	GC-MS			Dimethyl phthalate	1.6	100	30	46.9	75.8	36.9	165	18.2	88.9	87.8
37	GC-MS			Di-n-butyl phthalate	4.5	26	-	-	617	-	-	-	937	-
38	GC-MS		Other	Di(2-ethylhexyl)adipate	5	11	-	-	-	-	74.8	-	-	-
39	GC-MS	Domestic chemicals		3,5-di-tert-Butyl-4-hydroxybenzaldehyde	4.9	21	11.2	-	-	-	-	-	28.4	-
40	GC-MS			2-Methylphenol	1.95	5	-	-	-	5.79	-	-	-	-
41	GC-MS			Phenol	1.5	16	-	-	46.4	-	-	-	43.1	-
42	GC-MS			Butanoic acid, butyl ester	2.3	5	116	-	-	-	-	-	-	-
43	GC-MS			Bisphenol A	3.3	21	-	-	-	-	-	-	34	-
44	GC-MS			2-(Methylthio)-benzothiazol	2.7	32	21	-	-	-	13.8	-	-	-
45	GC-MS			2-Methoxyphenol	1.3	5	-	-	-	-	-	-	-	-
46	GC-MS			Cyclohexanol	1.2	5	-	-	-	-	-	-	-	-
47	LC-QTOFMS			Cotinine	0.2	5	-	-	-	-	-	-	-	-
48	GC-MS			Caffeine	0.07	21	-	-	-	-	-	58.8	-	220
49	LC-QTOFMS			Sucralose	1.4	5	-	-	-	-	-	1090	-	-
50	GC-MS	Industrial chemicals		Isophorone	1.7	21	-	-	-	-	-	90.4	-	-
51	GC-MS			Benzyl alcohol	1.1	11	7.77	-	-	-	-	-	-	-
52	GC-MS			N-Nitrosomorpholine	0.3	5	-	-	-	-	-	-	-	-
53	GC-MS			e-Caprolactam	0.12	5	-	-	-	-	-	-	-	-
54	GC-MS			n-Butylacrylate	2.4	47	2600	44.4	141	32	-	197	-	171
55	GC-MS			2-Naphthol	2.8	5	-	-	-	-	-	-	-	-
56	GC-MS			Nitrobenzene	1.8	5	-	-	-	7.43	-	-	-	-
57	LC-QTOFMS			N,N-Dicyclohexylurea	2.5	5	-	-	-	-	-	-	-	-
58	LC-QTOFMS			1H-Benzotriazole	1.3	5	-	-	-	-	-	-	-	-

No.	Detector	Origin	Group	Name	Log P	DF %	Suburban			Urban				
							G1	G2	G3	G4	G5	G6	G7	G8
59	LC-QTOFMS			Melamine	1.14	5	-	-	-	-	-	-	-	
60	GC-MS			Pentamethylbenzene	4.9	42	99.1	-	-	-	-	103	-	21.4
				Total concentration			5110	201	1370	296	545	1770	2480	2810
				Sum of pesticides			1710	-	-	57.4	0.8	20.1	2.90	1970
				Sum of sterols			-	-	53.9	-	-	66.7	612	-
				Sum of PPCPs			370	82	237	157	56.3	129	19.1	191
				Sum of FRs			143	27.7	200	-	219	-	334	146
				Sum of plasticizers			30	46.9	693	36.9	255	18.2	1400	88

DF: Detected frequency

RfD: Reference dose ($\text{mg kg}^{-1} \text{ day}^{-1}$)

RQ: risk quotient

RQ_{max} : maximum RQ value of individual compound

PPCPs: Pharmaceuticals and personal care products

<MDL: Below method detection limit

* "-" is not detected or toxicity data is not available

** : maximum total RQs

^a The RfD value was calculated with no observed adverse effect level (NOAEL) and lowest observed adverse effect level (LOAEL) by (Li et al. 2020)

^b The RfD value was predicted from the quantitative structure and activity relationship (QSAR) model with the conditional toxicity value (CTV) software (Wig)

^c From US The Risk Assessment Information System (RAIS) (RAIS 2024)

^d From US EPA Integrated Risk Information System (IRIS) (IRIS 2025)

3.3. Pesticides

Nineteen pesticides, including six insecticides, seven fungicides, five herbicides, and one plant growth regulator, were detected in 9 of the 19 groundwater wells (Table 1). To the best of our knowledge, these pesticides were reported for the first time in Vietnamese groundwater, with the majority identified using LC-QTOFMS. Pesticides were present in all urban wells, in contrast to the 33% detection frequency in agricultural areas, and similarly low levels in suburban and industrial areas. This highlights the widespread use of pesticides at urban sites, which is probably related to pest, fungal, and weed control.

The elevated total pesticide concentration occurred at G10 ($2,340 \text{ ng L}^{-1}$), G8 ($1,970 \text{ ng L}^{-1}$), and G1 ($1,710 \text{ ng L}^{-1}$), which were dominated by 3-hydroxycarbofuran. This compound is a metabolite of carbofuran and was shown to be more hydrophilic than that of its parent chemical but with equal toxicity (Mustapha et al. 2019). With a low log P of 2.32, carbofuran and its metabolites are highly mobile in soil and water (Mustapha et al. 2019), facilitating groundwater contamination via percolation (Mackay and Roberts, 1985). Carbofuran was commonly used to control pests in crops, gardens, and homes. Due to their harmful effects, carbofuran and other carbamate pesticides were banned in Vietnam since 2017 (Giao et al. 2021). However, the detection of 3-hydroxycarbofuran and furathiocarb may indicate the continued use of carbamate pesticides in Vietnam.

Contrastingly, organochlorine pesticides, reported earlier in urban groundwater in Vietnam in 2014 (Duong et al. 2015a), were not observed here. This is probably due to the ban on these pesticides in Vietnam in recent years (Le et al. 2023).

3.4. PPCPs and domestic-use chemicals

PPCPs were detected in 18 of the 19 wells, and seven compounds were identified. The number of PPCPs detected per well ranged from 0–5, with a median of 2. Total PPCPs concentrations (ΣPPCPs) varied from < MDL to 728 ng L^{-1} , with a median of 187 ng L^{-1} . All agricultural wells exhibited ΣPPCPs above 100 ng L^{-1} , with the maximum ΣPPCPs reached 728 ng L^{-1} at G10. Furthermore, ultraviolet filters, including ethylhexyl methoxycinnamate and octocrylene, are frequently found in agricultural wells, which may indicate their common use as sunscreens in rural communities.

The most frequently detected PPCP was 2-ethyl-1-hexanol followed by diethyltoluamide (DEET) with DF of 98% and 58%, respectively. Their frequent occurrence in groundwater is consistent with that of the global findings (Barnes et al. 2008; Duong et al. 2015a; Kong et al. 2016). As a versatile chemical, 2-ethyl-1-hexanol is widely used in consumer products and DEET is widely used as an insect repellent. Additionally, these chemicals were widely observed in Vietnamese surface waters (Chau et al. 2018).

Four phenolic compounds, phenol, 2-methylphenol, bisphenol A, and 2-methoxyphenol, were detected in the groundwater samples. Phenol, 2-methylphenol, and bisphenol A (BPA) are known or suspected endocrine-disrupting toxic chemicals (Jin et al. 2004; Kadoma et al. 2009; PubChem 2024). BPA, a widely recognized endocrine disruptor, was detected at concentrations up to 191 ng L^{-1} at G11. However, this level remain considerably lower than those found in Europe ($2,300 \text{ ng L}^{-1}$) (Loos et al. 2010) and the United States ($2,550 \text{ ng L}^{-1}$) (Barnes et al. 2008), and well below the short-term exposure threshold of $100 \text{ } \mu\text{g L}^{-1}$ (MDH 2013). Other phenolic compounds were detected only at trace levels.

Furthermore, DEET, BPA, caffeine, and cotinine are commonly used as molecular markers of wastewater pollution (Lamastra et al. 2016). Thus, their presence in the groundwater samples may indicate the influence of sewage and wastewater treatment plant discharge on groundwater pollution (Lamastra et al. 2016).

3.5. Plasticizers and fire retardants

Plasticizers were consistently detected in the groundwater samples, reflecting their common use in Vietnam. Five plasticizers were identified: four phthalates and di(2-ethylhexyl)adipate. Dimethyl phthalate (DMP) was the most ubiquitous plasticizer, with DF of 100% and median concentration of 113 ng L^{-1} . With its widespread application in plastics, inks, and repellents, DMP is frequently detected in surface water and groundwater globally (Dueñas-Moreno et al. 2022).

Di-iso-butyl phthalate (DiBP) and di-n-butyl phthalate (DnBP) occur at levels above 260 ng L^{-1} , particularly in agricultural areas where inputs may be derived from greenhouse plastic films, irrigation pipes, or agrochemical containers (Dueñas-Moreno et al. 2022). These findings indicate that agricultural practices may be a significant source of plasticizer contamination in Vietnam. Additionally, at G15, located near a river receiving industrial effluent, DnBP was detected at a concentration of $10,200 \text{ ng L}^{-1}$. The elevated contamination of DnBP at G15 may be related to industrial discharge near the study site. Although DiBP and DnBP were restricted in the United States and European Union because of their endocrine-disrupting and reproductive effects (Koch et al. 2012), these chemicals remain unregulated in Vietnam, raising concerns over chronic exposure risks.

Organophosphate esters were detected in 13 of 19 samples: tris (1-chloro-2-propyl)phosphate (TCPP), tri-n-butyl phosphate (TnBP), tri-iso-butyl phosphate (TiBP), and triphenyl phosphate (TPP). These chemicals are commonly used in plastics, foams, textiles, electronics, lubricants, and hydraulic fluids (Koch et al. 2012; Truong et al. 2023; Zweifel and Amos 2001); therefore, they are frequently detected in groundwater globally (Kong et al. 2016; Plantak et al. 2023; Regnery et al. 2011). This study is the first to report the presence of TCPP, TiBP, and TPP in groundwater in Vietnam, whereas previous research identified only TnBP (Duong et al. 2015a). This difference may be attributed to the application of both GC-MS and LC-QTOFMS analyses in the current study and the shifts in chemical consumption over the last decade. FRs are more frequently distributed in groundwater outside industrial zones, indicating that they may originate from diffuse sources, such as consumer products, building materials, or surface runoff (Koch et al. 2012; Truong et al. 2023; Zweifel and Amos 2001), rather than industrial discharge.

3.6. Possible sources and pathways

Groundwater contamination by OMPs is strongly influenced by compound properties, such as solubility and persistence (Mackay and Roberts 1985). Water-soluble chemicals are frequently detected in aquifers because they can readily percolate (Mackay and Roberts 1985). However, OMPs with high ($\log P < 1$) and moderate ($\log P: 1-3$) water solubilities (Schwarzenbach et al. 2016) accounted for only 10% and 45% of the detected compounds, respectively. Hydrophobic compounds ($\log P > 3$) (Schwarzenbach et al. 2016) accounted for 45% of the detected OMPs. As hydrophobic OMPs are typically retained in soils and sediments, numerous studies in Vietnam report their accumulation in soils and sediments at $\text{ng-}\mu\text{g kg}^{-1}$ levels (Jayasekara et al. 2025; Matsukami et al. 2017; Nishina et al. 2010; Trinh et al. 2020). Over time, they may gradual migration into aquifers (Dueñas-Moreno et al. 2022), i.e., the more hydrophobic the OMPs more the retardation (Mackay and Roberts 1985).

Furthermore, borehole leakage is considered an important pathway for groundwater contamination by OMPs in the study areas. This explanation was supported by the detection of several hydrophobic but low-persistence compounds, such as cyprodinil, 2-ethyl-1-hexanol and phenol (PubChem 2024), suggesting alternative transport routes bypassing natural sorption barriers and reducing retardation. As the study areas are located in the Red River alluvial plain, where pore-water aquifers dominate (Bui et al. 2011), transport via fractures or karst conduits is unlikely. Instead, leaky boreholes emerged as a possible pathway because poorly constructed or abandoned wells can serve as conduits for contaminants to migrate into deeper aquifer layers (Jiménez-Martínez et al. 2011). In Vietnam, the use of household boreholes were common (Tran et al. 2016), however, earlier surveys showed that $\sim 20\%$ of household wells were located near septic tanks or waste sites, and many exhibited casing, cap or courtyard cracks allowing pollutant intrusion (Tran et al. 2016n et al. 2023). Furthermore, structural deterioration commonly occurs in aging boreholes, particularly those with metal casings (Thanh and Tung 2018), which further increases their vulnerability to contamination. Given that all the wells surveyed here have been in operation for more than a decade, they may exhibit a high likelihood of structural deterioration. Additionally, an earlier reported high coliform levels in well water linked to wastewater leakage (Duong et al. 2011). Such borehole pathways are particularly critical during rainy seasons, when runoff can accelerate pollutant entry into deteriorated or poorly sealed wells.

Beyond the physicochemical properties, the environmental half-life of OMPs in water can provide further information on whether the detected OMPs represent legacy residues or recent inputs. Compounds with long half-lives in water environments (weeks to months), such as organophosphate esters (TCPP, TiBP and TPP), plasticizers [di(2-ethylhexyl) adipate and BBP], melamine, and some pesticides (carbendazim, fenazaquin, atrazine, and diuron) (PubChem 2024), can persist in aquifers, reflecting historical or continuous contamination sources. Contrastingly, the presence of short-lived compounds with reported half-lives of only days suggests that they originated from continuous or recent sources. The coexistence of persistent and short-lived OMPs in Vietnamese aquifers reflects a combination of legacy contamination and present-day pollution sources.

To explore the similarities in OMP contamination profiles, PCA was performed on 19 groundwater samples using the concentrations of 21 OMPs with DF > 20% (Fig. S1). The first (PC1) second (PC2) components explained 28.1% and 49.4%, respectively.

Three clusters were identified here. Cluster 1 grouped G17–G19 (industrial), G4 (urban), and G2 (suburban), and was characterized by high PC1 loadings (> 0.75). This suggests that industrial sites, together with adjacent urban and suburban areas, share contamination sources likely linked to wastewater and industrial discharge. Cluster 2 consisted mainly of G11–G14 (agricultural), together with urban sites G5, G6, G8, and suburban site G1, all of which showed similar loadings for both PCs. This cluster highlighted the mixing of agricultural and domestic signatures, which likely reflects agricultural diffusion combined with municipal inputs. Cluster 3 included G9, G10 (agricultural), G7 (urban), G15 (industrial), and suburban G3, as defined by high PC2 loadings. Their grouping points to cross-boundary contamination, where agricultural, industrial, and urban sources converge in shared hydrogeological settings.

Overall, PCA revealed that OMP profiles were influenced not only by local land use but also by overlapping source contributions and transport across boundaries. The observed similarities probably resulted from the comparable chemical usage patterns, preferential flow pathways, and shared hydrogeological conditions of the Red River delta aquifers. However, the exact routes of introduction and contamination mechanisms of OMPs in Vietnamese aquifers require further investigation.

3.7. Risk assessment

Not all the surveyed wells served as primary sources of drinking water; however, according to a site survey, these wells provide water for domestic use, and many families still rely on well water as a secondary drinking water source. A significant portion of the water supply of Hanoi comes from groundwater (Duong et al. 2015a). Therefore, we performed a health risk assessment of OMPs in groundwater under the assumption that groundwater serves as a drinking water source. As toxicity data were not available for all the detected compounds at the time of this study, a human health risk assessment was performed for only 25 OMPs with available toxicity data (Table 1 and Table S9).

The health risk assessment of 25 OMPs in groundwater revealed that all RQs values were much lower than 1, with DnBP exhibiting a maximum RQ of 0.003. These results suggest that the observed concentrations of OMPs in groundwater samples from Vietnam are unlikely to pose significant health risks to residents, even those relying on well water as a drinking water source.

To account for the combined toxicity of OMPs, the total RQs of the 25 calculated compounds were used to account for the combined toxicity of OMPs. The highest total RQ of 0.003 at G15 remained far below 1, indicating that the combined effects of these 25 OMPs are not expected to threaten human health.

However, we did not assess the health risk for all the detected OMPs due to the lack of toxicity data. Thus, further investigations on the impact of OMPs in groundwater, along with periodic monitoring of the occurrence of OMPs in well water, should be undertaken to obtain more comprehensive information on the behavior and impact of OMPs in groundwater on resident health.

Conclusions

This study confirmed the reliability of the travel analysis method, enabling its application to international sample investigations. The measured results of groundwater samples in Vietnam revealed 1) the occurrence of 60 OMPs, including ~ 40 compounds reported for the first time in Vietnamese groundwater. 2) Land-use specific contamination patterns were identified, with pharmaceuticals and plasticizers dominating agricultural wells, and fire retardants, pesticides, and household-use chemicals being prevalent in urban sites. 3) Urban groundwater OMP profiles have changed over the past decade, and Vietnamese aquifers now face both legacy and ongoing pollution. 4) Beyond migration from surface sources to aquifers via percolation, leaky boreholes may be a possible pathway for surface contaminants to enter aquifers in Vietnam. 5) Calculated individual and cumulative risk quotients suggested no immediate human health risks. Although this survey did not identify serious risks, various OMPs exist widely, highlighting the need for monitoring systems, zone-specific preventive management strategies and national regulations to ensure sustainable groundwater use in developing Asian countries.

Declarations

Competing Interests

The authors have no relevant financial or non-financial interests to disclose.

Author Contributions

All authors contributed to the study conception and design. Dung Anh Truong: Investigation, Formal analysis, Data treatment, Writing (Original Draft), Writing (Review & Editing). Takashi Miyawaki: Supervision, Writing (Review & Editing). Hanh Thi Duong, Ha Thu Trinh, Takumi Goto and Nguyen Thi Thanh Hai: Investigation, Formal analysis. Hidenari Yasui: Review. All authors read and approved the final manuscript.

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Author Contribution

All authors contributed to the study conception and design. Dung Anh Truong: Investigation, Formal analysis, Data treatment, Writing (Original Draft), Writing (Review & Editing). Takashi Miyawaki: Supervision, Writing (Review & Editing). Hanh Thi Duong, Ha Thu Trinh, Takumi Goto and Nguyen Thi Thanh Hai: Investigation, Formal analysis. Hidenari Yasui: Review. All authors read and approved the final manuscript.

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Figures

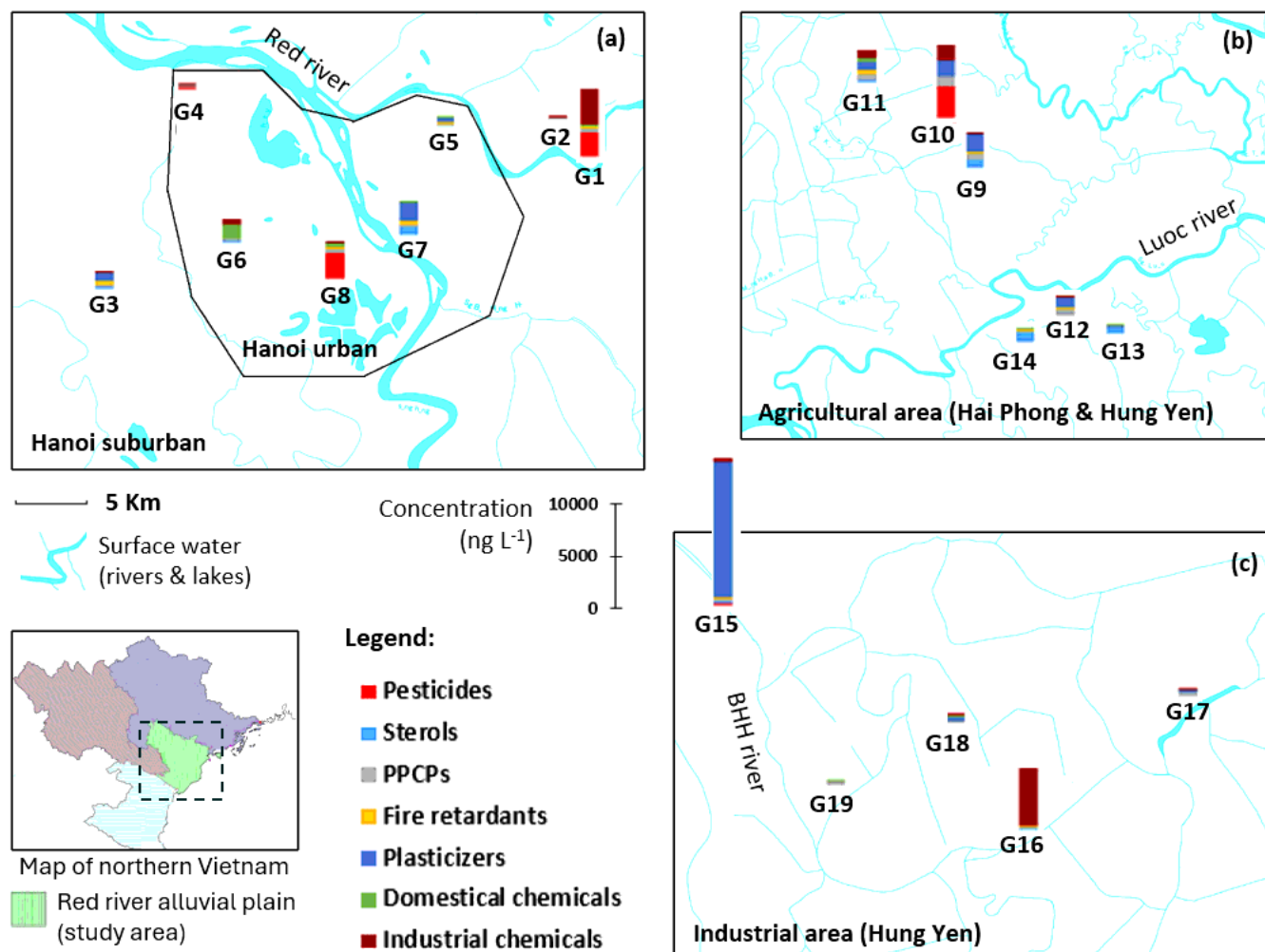


Figure 1

Spatial distribution of OMP concentrations in sampling wells in Vietnam: (a) urban and suburban areas, (b) agricultural area, and (c) industrial area

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