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Natural Attenuation of Arsenic in Natural Wetlands at Thung Kham Gold Mine Wang Saphung District, Loei Province

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Abstract

The gold mining operations in Khao Luang Sub-district, Wang Saphung District, Loei Province, have had a significant impact on the surrounding environment, leading to arsenic contamination. The objective of this research was to examine the extent of arsenic contamination in the natural wetland area and Pu Leuk Creek, adjacent to the Thung Kam gold mine. The study focused on sediments samples taken from three points along the creek: upstream, middle, and downstream, at depths ranging from 0 to 150 cm. The findings revealed that concentrations of total arsenic in the sediment at the upstream, middle, and downstream points ranged from 160.1-1,112 mg/kg, 49.79-1,911 mg/kg, and 0.39-1,080 mg/kg, respectively. For As(III), the concentrations ranged from 28.63-320.40 mg/kg, 0-1,032 mg/kg, and 0-544.60 mg/kg, respectively. For As(V), the concentrations ranged from 115.49–853.70 mg/kg, 8.5-879 mg/kg, and 0–586.5 mg/kg, respectively. Most of these values exceed the standard threshold for soil quality for agricultural use (25.00 mg/kg) set by the National Environmental Board in 2021. The highest concentrations of arsenic were found at the middle point of the creek, possibly due to its proximity to the mineral waste pond in the gold mining area. Additionally, the highest concentrations of arsenic were found at depths of 0–40 cm, indicating that depth levels affect the accumulation of arsenic in the sediment. The analysis of the elemental composition in the sediment using the Energy Dispersive X-ray Fluorescence (EDXRF) technique revealed that the chemical constituents in the sediment include Al, Si, K, Ca, Ti, Mn, Fe, Cu, Zn, As, Rb, Zr, and Pb. The predominant mineralogical components in the sediment are quartz (SiO_2) and hematite (Fe_2O_3). Regarding the analysis of the forms of arsenic in the sediment, it was observed that the concentration of As(V) was higher than that of As(III). This suggests that As(V) may have been absorbed or precipitated along with other mineral elements in the sediments. The presence of arsenic exceeding the standard limits in this area, as mentioned above, may have adverse health effects on the people residing near the gold mining area.

Keywords : Arsenic; Gold mine; heavy metal; Loei

Introduction

In nature, arsenic is considered a highly toxic semimetal, primarily found in the forms of arsenite (As(III)) and arsenate (As(V)). Arsenic predominates in its +5-oxidation state, As(V), in the form of arsenate oxyanions ($H_2AsO_4^-$, $HAsO_4^{2-}$). The high affinity of As(V) for Fe(III) (oxyhydr) oxides may effectively immobilize aqueous its [1-4]. Arsenite (As(III)) is more toxic than arsenate (As(V)) due to its higher ability to penetrate cells and disrupt cellular processes. Both forms pose significant health risks, including cancer and organ damage, with arsenite being more mobile and readily absorbed by organisms. Effective treatment strategies, such as oxidation and adsorption, are necessary to manage both forms and reduce their harmful effects on health and the environment. In Thailand, arsenic occurs naturally throughout the country, mostly on the western side, such as in Suphan Buri Province, and has been discovered through excavation in provinces like Nakhon Si Thammarat, Phichit, and Loei. Arsenic contamination in the environment can stem from two main sources: natural resources and human activities. Natural contamination is associated with the weathering and degradation of rocks or minerals that store arsenic [5]. Human-induced contamination arises from activities such as mining, industrial waste disposal, leaching of arsenic-containing minerals, combustion of fossil fuels, and the use of arsenic in various products, such as pesticides [6]. Generally, naturally occurring arsenic is found within mineral pathways, often in conjunction with other compounds such as copper, iron, manganese, cobalt, nickel, silver, and gold. Therefore, mining activities may disperse arsenic into the surrounding areas [7]. In wetland environments, arsenic is retained mostly in sediments or media. Arsenic in wetlands can precipitate and form insoluble sulfide compounds or arsenopyrite (FeAsS) [8-11]. Coprecipitation processes can obtain high removal efficiency in the presence of sulfate, ferric chloride, and Iron oxide [11, 12].

The gold mining activities of Thung Kam Company in Wang Saphung District, Loei Province, have been identified as the root cause of arsenic contamination in the surrounding areas between the years 2014-2018. It was found that the highest accumulation of arsenic was in the soil samples and water sources near the gold mining area, resulting from leaching and absorption on the lateritic soil rich in iron and manganese. This situation has impacted the health of the residents living near the mining site. Investigations revealed that people's bodies were contaminated with arsenic, cadmium, lead, manganese, and cyanide. Moreover, elevated levels of arsenic were found in both water sources and soil samples, exceeding the standard limit of 0.01 mg/L. This conflict has led affected individuals to mobilize collectively through community-based governance, demanding government intervention to address the issue [13].

Additionally, assessments of water quality and heavy metal contamination in sediment, fish, and frogs, including the bioaccumulation factor (BAFs) observed in these aquatic organisms, have revealed concentrations surpassing regulatory limits [15]. Notably, arsenic in the sediment exists predominantly in the forms of arsenate (As(V)) and arsenite (As(III)), with arsenite being more mobile and toxic than As(V). Various soil elemental minerals have also been found to influence the transformation and species of Arsenic, as well as its storage and movement in the environment. Consequently, our research aims to investigate the extent of arsenic contamination in the natural wetlands and Phu Lek Creek adjacent to the Thung Kam gold mine. This investigation focused on analyzing dispersion patterns over distance and depth, as well as the chemical components influencing arsenic dispersion patterns.

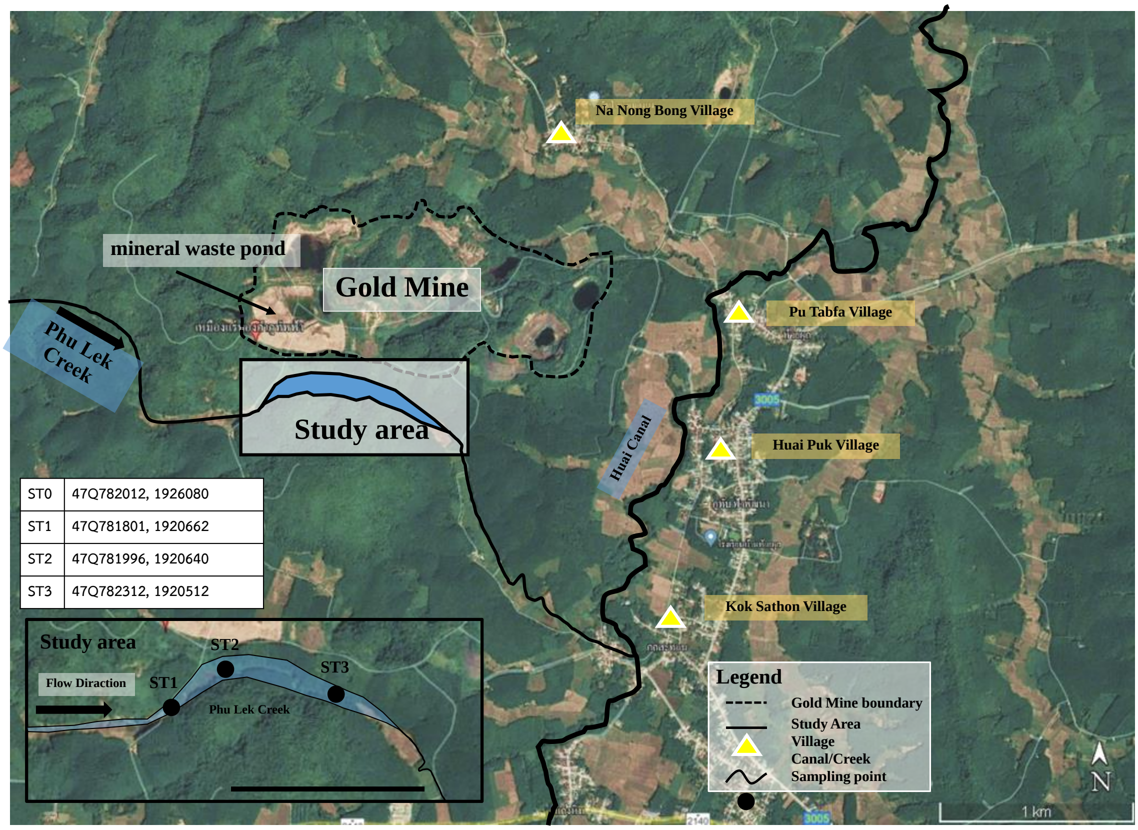
Methodology

Study area

The natural wetland area is located along Phu Lek Creek adjacent to the mineral waste pond at the end of the gold mine (which is currently closed). The gold mine operated as an open-pit mine and ore-dressing plant from the year 2006 to 2013. Water from the Phu Lek Creek flows downstream from the area behind the mine, sometimes receiving wastewater from the waste pond. The natural wetland receives water from two sources: wastewater from the mine to the northwest and underground springs from the northeast. The wetland area extends approximately 643 meters in length, with a width ranging from 10 to 50 meters and a depth of 0.20 to 1.0 meters. The total study area is approximately 10,000 sq.m. (Figure 1).

Sampling and preparation of sediment samples

In this study, sediment core samples were collected at sampling points along the creek, as illustrated in Figure 2 (1-10 March 2023, dry season). The samples were collected at a depth of 150 cm., with 5 cm. intervals at three points along the distance: the upstream at 0 m., middle at 290 m., and the downstream at 640 m. They adjusted their conditions by air-drying them in a shaded area for 7-14 days, depending on the soil moisture content. After that, the samples were heated at 105 degrees Celsius for 24 hours. Subsequently, the dried samples were ground using a ceramic mortar and passed through a 2-millimeter sieve in preparation for further analysis.



ST0	47Q782012, 1926080
ST1	47Q781801, 1920662
ST2	47Q781996, 1920640
ST3	47Q782312, 1920512

Figure 1 Map of the gold mine and Phu Lek Creek


(a) Upstream

(b) Middle

(c) Downstream

Figure 2 Sampling point characteristics: (a) Upstream (b) Middle and (c) Downstream

Sediments preparation

Sediments were collected from 3 sampling points along Phu Lek Creek, where the location was illustrated in Figure 1. Samples were classified according to particle size by sieving (all sizes, < 2 mm and > 2.00 mm). After the sieve, the samples were air-dried at room temperature. Sediment was analyzed for chemical and physical properties as shown in Table 1.

Physical characteristics: The analysis of sediment texture by hydrometer shows that the sediment sample is composed of sand 33.78%, silt 23.22%, and clay 43%. The high percentage of clay appears as a fine texture with low permeability.

Chemical properties: Sediments are mainly composed of Quartz (SiO_2), Hematite (Fe_2O_3), and Bauxite ($Al_2O_3.H_2O$). The EDX study and the X-ray diffraction spectrum of sediments indicated that the elemental compositions are Mg, Al, Si, S, K, Ti, Fe, and As (Table 1). From these results, it can conclude that sediment is composed of a high level of aluminum ($Al_2O_3.H_2O$ and $AlSi_2O_6$ (OH_2)) and iron oxide (Hematite). Physical appearance studied by SEM indicated that all sediment samples had low porous and rough surfaces which occurred from a binding of a small particle on the surface soil to generate shallow porous.

The pH of all sediment samples is between 4.76-5.04 which is considered a weak acid (Land Development Department, 2010). The electrical conductivity of the samples is lower than 0.1 ds/m which is considered as non-saline soil. Organic matters in all samples were moderate (1.5-2.5). The amount of iron was found highest in soil size < 2 mm 134,300 mg/kg (Table 1).

Analysis of total arsenic, As(III) and As(V)

Sediment samples were prepared in accordance with US EPA test methods for solid waste evaluation. Physical/Chemical Method (SW-846). Initially, 0.15 g of each dried sample was placed in a beaker and digested using a hot plate with a mixed acid concentration of 10 mL of HNO_3 and 5 mL of HCl. After cooling to room temperature, the suspended material was filtered through 0.42-µm filters. The solution was then diluted with deionized water. The digested samples were determined for arsenic using inductively coupled plasma optical emission spectrometry (ICP-OES, Agilent 5800 series, Santa Clara, CA, USA). For the analysis of As(III) and As(V), the ion exchange resin technique was used according to the methods specified in the American Public Health Association standards (APHA, 2012).

Energy Dispersive X-Ray Fluorescence (EDXRF)

The middle-point sediment, which showed the highest concentration of arsenic, was dried at 105 degrees Celsius and finely ground to prepare for elemental analysis. The ground sediment samples, with three replicates, were analyzed using the EDXRF spectrometer technique (Horiba XGT-5200). The power specifications of the tube are 50 kV maximum and 1 mA. The selection of filters, tube voltage, sample position, and current were fully computer-controlled, with an energy range of 0–40 keV. Quantitative analysis was conducted using the built-in software.

Table 1 Sediment characteristics and analytical methods

Parameters;		Particle size		Methods/ reference
	All size	**> 2 mm**	**< 2 mm**	
Physical properties				
Particle size (mm)		0.002 – 2.00		Sieve analysis, SEM-EDX, Carter and Gregorich (2006)
Bulk density (g/cm^2)	2.01	2.55	1.75	Core method, Carter and Gregorich (2006)
Surface area (m^2/g)	17.03	18.15	16.00	BET analysis (2013)

Parameters;		**Particle size**		**Methods/ reference**
	All size	**> 2 mm**	**< 2 mm**	
Chemical properties				
pH	5.10	4.76	5.04	1:5, Sediments:water mixture, pH meter, APHA (2012)
pH_{ZPC}	5.80	4.75	4.91	1:5, laterite:water mixture, pH meter, APHA (2012)
EC (ds/m)	0.025	0.036	0.021	1:5, Sediments:water mixture, EC meter, APHA (2012)
CEC (c mol(+)/kg)	25.14	23.36	33.45	Ammonium acetate extraction (1N) Land Development Department, (2010)
Organic matter (%)	2.08	2.17	1.96	UV254, APHA (2012)
Inorganic composition (by SEM-EDX)				SEM-EDX, model: ESM-5800, GEOL, Japan
Magnesium(Mg) (%)	0.44	0.39	0.15	
Aluminum(Al) (%)	24.84	22.54	24.47	
Silicon(Si) (%)	44.01	44.68	42.55	
Sulfur(S) (%)	0.14	0.17	0.43	
Potassium(K) (%)	2.76	2.72	2.56	
Titanium(Ti) (%)	1.44	1.41	1.41	
Iron(Fe) (%)	26.18	27.93	28.39	
Arsenic (As) (%)	0.19	0.16	0.05	
Metals (by ICP-OES)				Digestion with 1:3(HNO_3 : $HClO_4$)(v/v), ICP-OES, Perkin Elmer, Optima 8000 APHA (1998)
Iron(Fe) (mg/kg)	99,640	98,030	134,300	
Arsenic (As) (mg/kg)	0.17	0.13	0.16	

Statistical analysis

Statistical analysis was performed by the SPSS statistics 17.0 for Windows software package program network Licensed in Suranaree University of Technology. Data was calculated by mean, minimum, maximum, and standard deviation for a result. The significance of the difference between depth and arsenic species was determined with a *t*-test (Significant at a level of 0.05). Quality assurance (QA) and quality control (QC) were used in planning, sampling, analysis, and reporting of data in all processes throughout the study. Pearson correlation coefficient (significant at a level of 0.05) was used to determine the correlation between date, sediment depth, and arsenic (total/As^{3+}/As^{5+}).

Results and Discussion

Sediment properties in the wetland

The results of the analysis of sediment properties are shown in Table 1, which found that most of the analysis results have values consistent with laterite soil properties [14], namely Particle size (0.025-2.00 mm.), Bulk density (1.98-2.32 g/cm^3), Surface area (15.52-18.35 m^2/g), Pore volume (0.011-0.020 ml/g), Organic Matter (0.56-2.08%), Iron (Fe) (27.93-28.39%) etc.

Study of total arsenic, As(III) and As(V)

The concentrations of total arsenic in sediment at the upstream, middle, and downstream at depths of 1-150 cm. ranges from 160.1-1,112 mg/kg, 49.79-1,911 mg/kg, and 0.39-1,080 mg/kg, respectively. (Table 2) For As(III), the concentrations range from 28.63-320.40 mg/kg, 0-1,032 mg/kg, and 0-544.60 mg/kg, respectively. As for As(V), the concentrations range from 115.49-853.70 mg/kg, 8.5-879 mg/kg, and 0-586.5 mg/kg, respectively. Most of the values exceed the standard threshold for soil quality for agricultural use (25.00 mg/kg) set by the National Environmental Board in 2021.

Figure 3, which illustrates the relationship between depth and the total amount of arsenic at the Upstream point, Middle, and Downstream, it is evident that as depth increases, the quantity of arsenic in the sediments decreases. Additionally, the highest concentration of arsenic was found at depths of 0-40 cm., indicating that depth levels influence the accumulation of arsenic in the sediments. Furthermore, at depths of 0-40 cm., conducive to root growth, arsenic tends to be trapped on plant roots and accumulates predominantly in the sediments. Moreover, the highest arsenic concentration was observed around the middle of the stream, possibly due to its proximity to the mine tailings in the gold mining area.

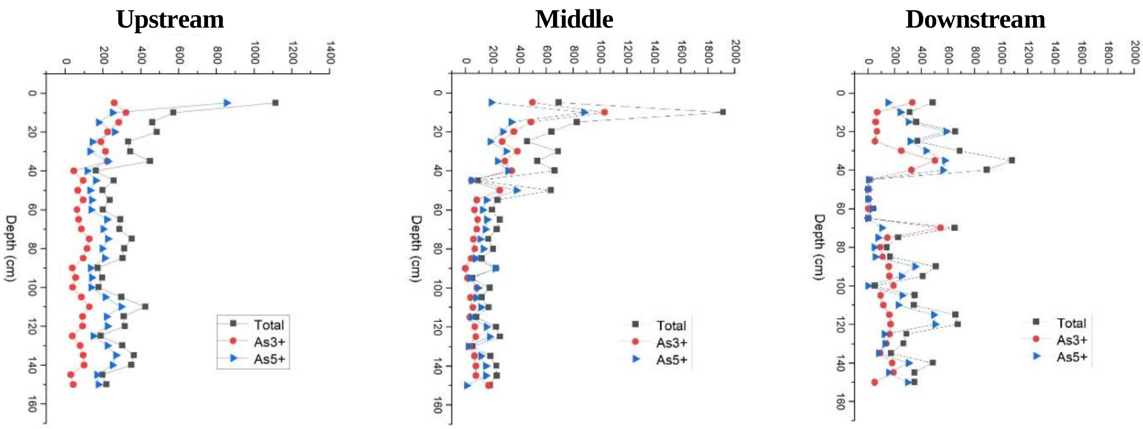
Figure 4, which illustrates the relationship between As(III) and As(V), shows that at higher concentrations, particularly at depths of 0–40 cm, arsenic is predominantly found in the form of As(V) rather than As(III). This is because As(V) is more stable and exists in oxidizing environments. As the depth increases, arsenic is mainly present in the form of As(III) and is found at lower concentrations compared to As(V). At greater depths, arsenic in pore water diffuses into a reducing environment, leading to in-situ precipitation into stable forms, As(V) [16], resulting in higher concentrations of As(V) than As(III). In areas with a high amount of Fe(III) (ferric oxyhydroxide), arsenic may oxidize to form arsenopyrite (FeAsS) and accumulate in the soil [4, 14].

Table 1 Sediment properties

Properties	**Quantitative value**
Particle size (mm)	0.021-2.10
Bulk density (g/cm^3)	1.18-2.20
Surface area (m^2/g)	13.85-18.00
Pore volume (ml/g)	0.013-0.020
pH$_{ZPC}$ (1:5, laterite: water mixture)	5.30-7.10
Conductivity (1:5, laterite: water mixture) (µS/cm)	40.00-52.60
Organic Matter (%)	0.86-2.080
Magnesium(Mg) (%)	0.17-0.40
Aluminum(Al) (%)	22.40-24.40
Silicon(Si) (%)	42.20-45.00
Sulfur(S) (%)	0.11-0.20
Potassium (K) (%)	1.06-2.00
Titanium (Ti) (%)	0.21-1.40
Iron(Fe) (%)	27.90-28.30

Table 2 Total arsenic, As(III) and As(V) content in natural wetlands

	Upstream			**Middle**			**Downstream**		
	Total As	**As(III)**	**As(V)**	**Total As**	**As(III)**	**As(V)**	**Total As**	**As(III)**	**As(V)**
count	30	30	30	30	30	30	30	30	30
Max (mg/kg)	1,112	320.4	853.7	1911	1032	879	1080	544.6	586.5
Min (mg/kg)	160.1	28.63	115.49	49.79	0	8.5	0.398	0	0


Upstream

Middle

Downstream

Figure 3 Concentration of total arsenic, As(III) and As(V) with depth

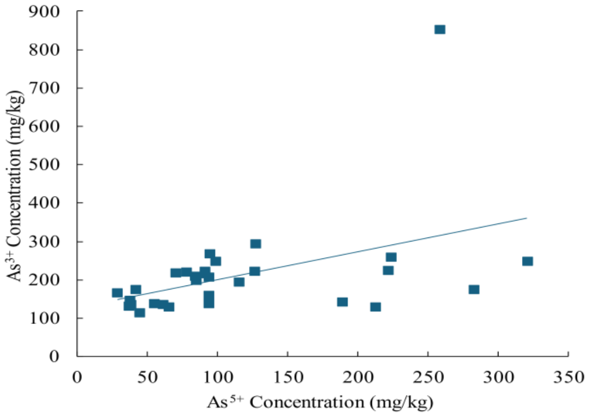


Figure 4 Difference between As(III) and As(V)

Results of the study of elemental composition in sediments

The analysis of elemental composition in the sediment using Energy Dispersive X-ray Fluorescence (EDXRF) technique revealed that the chemical constituents in the sediment consist of Al, Si, K, Ca, Ti, Mn, Fe, Cu, Zn, As, Rb, Zr, and Pb, as shown in Figure 5. The predominant mineralogical components in the soil sediment are quartz (SiO_2) and hematite (Fe_2O_3). Furthermore, Table 3 reveals that iron is found in higher quantities compared to other mineral elements in the sediments samples. This is attributed to the predominance of iron in the environmental conditions of the Thung Kam gold mine area [14].

Mechanisms of natural attenuation of arsenic in the wetland

In this study of the natural wetland area of the Thung Kam gold mine, the predominant mineralogical components in the sediment were quartz (SiO_2) and hematite (Fe_2O_3). The arsenic content accumulated in the sediments ranged from 0.39 to 1,080 mg/kg. In wetland environments, arsenic is primarily retained in sediments or media rather than accumulated in plants. Depending on redox conditions, arsenic in wetlands can precipitate and form insoluble sulfide compounds, such as arsenopyrite (FeAsS) [8-11]. Coprecipitation processes can achieve high removal efficiency in the presence of sulfate, ferric chloride, and iron oxides [11, 12].

In this study, the highest arsenic concentration was found at depths of 0–40 cm, where arsenic predominantly occurred in the form of As(V) rather than As(III). At greater depths, arsenic was mainly present in the form of As(III) and at lower concentrations compared to As(V). The sediment was rich in Fe(III) (ferric oxyhydroxide), which facilitated the oxidation of arsenic in water by Fe(III), leading to its adsorption or precipitation along with other mineral elements in the sediments. At depths less than 40 cm, which are conducive to root growth, arsenic tends to be trapped on plant roots and accumulates predominantly in the sediments.

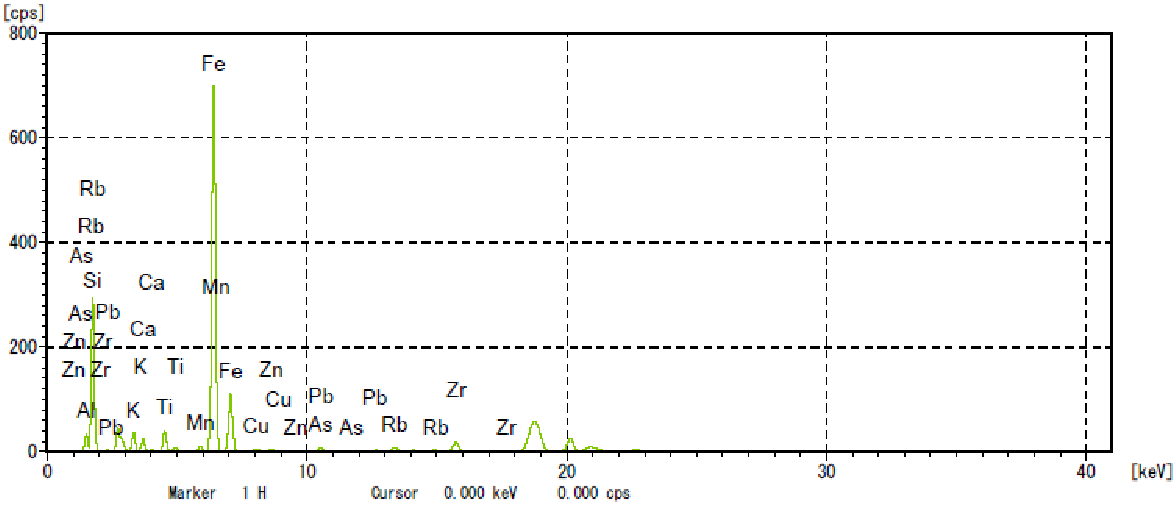


Figure 5 Chemical composition of sediments in natural wetlands

Table 3 Descriptive statistics of heavy metals in the sediment profiles in natural wetlands (%)

	Statistic	Fe	Al	Mn	Cr	Pb	As	P	Zn
Soil Profiles	Maximum	78.21	6.47	1.92	0.11	0.00	0.63	0.30	0.43
	minimum	31.61	0.00	0.35	0.03	0.00	0.21	0.00	0.10
	Mean	47.13±1.86	1.23±0.34	0.75±0.07	0.07±0.004	0.00	0.41±0.22	0.05±0.01	0.22±0.01
	SD	10.18	1.91	0.36	0.02	0.00	0.12	0.08	0.76

Conclusions

From the study of sediment at depths of 0-150 cm, it was found that the total arsenic, As(III), and As(V) concentrations exceeded soil quality standards for agricultural purposes. The depth level with the highest accumulation of arsenic was 0-40 cm, indicating that sediment depth influences arsenic accumulation. The main chemical constituents in the sediment in the wetland area are quartz (SiO_2) and hematite (Fe_2O_3). Regarding the analysis of the forms of arsenic in the sediment, it was observed that the concentration of As(V) was higher than As(III). This suggests that As(V) may have been absorbed or precipitated along with other mineral elements in the sediments.

The presence of arsenic exceeding standard limits in this area may pose significant health risks to residents near the gold mining site. The sediment in the Phu Lek wetland area is laterite, with iron oxides as its main component. This type of soil can remove arsenic from water with 50-93% efficiency [14]. The arsenic levels detected were far above standard, resulting in contamination of water, plants [17], and food [18] around the gold mining area. Therefore, mitigation and remediation measures for arsenic contamination, such as land improvement, soil rehabilitation, phytoremediation, and soil amendments, are recommended to reduce arsenic exposure. Furthermore, future research should focus on monitoring arsenic migration, assessing its bioavailability to crops, and exploring the health impacts on the local community to mitigate these risks more effectively.

Acknowledgements

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