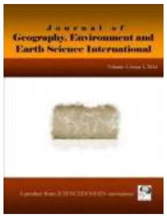

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Contamination of Agricultural Soils by Toxic Trace Metals in an Industrial District in Vietnam

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Authors' contributions

This work was carried out in collaboration between all authors. Author TNV designed the study, performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Author AO assisted the analyses of the study. Authors YX and AND assisted the field sampling and managed the literature searches. Author KK provided laboratory equipment in Japan and revised the final version of the manuscript. All authors read and approved the final manuscript.

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ABSTRACT

The contamination of agricultural soils by the toxic trace metals arsenic (As), lead (Pb), cadmium (Cd), chromium (Cr), copper (Cu) and zinc (Zn) was investigated in an industrial district in Vietnam. In this district, irrigation agriculture is conducted via channels; additionally, there are two industrial parks on nearby agricultural land. The purpose of the study was to clarify the magnitude and spatial distribution the trace metal concentrations, the source of the trace metals, the difference was in concentration between the two industrial park areas, and the usability of the soils for agriculture. In

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this study, eighteen soil samples were taken, and the trace toxic metals in soils were determined using atomic absorption spectrometer. Our results showed that the trace metal concentrations were in the order of Cr > Zn > Pb > Cu > As > Cd for the district. No significant differences were observed between the two park areas in terms of concentration. Cr and Zn concentrations were highest near the factories in the park areas, and the other trace metals were also comparatively high in these areas, demonstrating that the trace metals originated from the factories via wastewater. The correlations observed in the concentrations between the Cd, Pb and Zn, Cr and Zn, and Cu and As suggested that there were several sources of wastewater supplying the trace metals. Concentrations of all trace metals exceeded the permissible level for agricultural soils; therefore, remediation measures are necessary to reduce contamination.

Keywords: Toxic trace metals; soil contamination; industrial waste water; agricultural soils; irrigation water.

1. INTRODUCTION

The contamination of agricultural soils by trace metals from irrigation water mixed with industrial wastewater has been reported frequently since 2000. These reports have been made worldwide from diverse countries including China, Bangladesh, Egypt, Ghana, and India [1–6]. According to these reports, the dominant trace metal contaminant in the soils was different depending on area; however, the concentrations of the trace metals found mostly exceeded the permissible levels for agricultural soils, indicating a serious contamination [7,8]. Furthermore, it was shown that crops grown in contaminated soils were also contaminated with the trace metals [9–11]. Contamination of soils by trace metals can be caused by long-term irrigation, despite the irrigation water being uncontaminated with trace metals [5,12–14]. The trace metal concentrations of the soils were higher on the soil surface and were lower as the soil depth became deeper [15–17].

In Vietnam, soil trace metal contamination in Hanoi City has been reported as follows. Agricultural soils in a factory-concentrated area were contaminated with As, Cd, Cr, and Zn, caused by irrigation water contaminated with industrial waste [18,19]. According to Phuong et al. [20], soils from a paddy field located in a copper casting village were contaminated with Cu, Pb, and Zn. The agriculture soils not only near industrial zones but downstream areas of rivers were also affected by irrigation water that received wastewater from anthropogenic activities upstream [21–23].

Since 2000, industrial parks built outside Hanoi have discharged untreated industrial wastewater into nearby rivers. River water is used for crop irrigation in agricultural lands near factory areas, and the trace metal contamination is believed to occur in agricultural soils; however, the situation of the contamination is yet to be clarified.

Therefore, in this study, the trace metal contamination of agricultural soils was investigated in a district of Hung Yen Province located near Hanoi City, where two industrial parks are located, with different foundation years and factory composition. Irrigation for agriculture is performed through channels there. The present study investigated five toxic trace metals that potentially contaminated the soil, and was designed with the intention to clarify the magnitude and spatial distribution of the soil trace metal concentrations and the source of the trace metals through the difference in concentrations between the two industrial park areas, and the usability of soils for agricultural purposes, respectively.

2. MATERIALS AND METHODS

2.1 Study Area

The study area is an agricultural area belonging to the districts of Van Lam, Yen My and My Hao in Hung Yen Province, which neighbors Hanoi City. Two industrial parks, Pho Noi and Thang Long, are in the study area (referred to as the study district), whose areas are 6 and 2.2 km^2, respectively. The total population of these areas was 351,340 in 2013, occupying approximately 30% of the provincial population.

The Pho Noi industrial park was built in 2003. Forty-six percent of the factories are mechanical factories, 13% plastic factories, 5% electronic factories, and the remaining industries consist of chemical, textile, food, wood and ceramic factories. In contrast, the Thang Long industrial park was built in 2009 and 38% of the factories are mechanical, 14% plastic, 14% electronic manufacturing, and the remainder are steel, fluorescent lamp, and electric conductivity equipment factories. Wastewater from the factories is discharged into drains from where the drain water flows into irrigation canals.

According to the Asian Development Bank [24], the average air temperature in the district is 23℃ during summer and 16℃ during winter, and average annual humidity is 84%. Total annual rainfall is 1584 mm, and the total rainfall in the rainy season reaches 80–85% of the total annual rainfall. The rainy and dry seasons are May–October and November–April, respectively. The topography of Hung Yen Province is flat without hills and mountains, with many rivers where flooding is common in the rainy season.

2.2 Soil Sampling and Chemical Analysis

A total of 18 sites (points) in the district were targeted for soil sampling in paddy fields after considering the distribution of the irrigation canals. The sampling was done in August, 2013 and the locations of the sampling are shown in Fig. 1. The soil collection was done at 5 spots for the respective points. At one spot, 1 kg topsoil (0–20 cm) was collected by using a stainless-steel drill. The soils were stored in plastic bags until use. All soils were air dried, crushed using a ceramic mortar and sieved through a 2 mm sieve to remove gravels and plant residues. Five topsoil samples were thoroughly mixed to be homogeneous. before 500 g soil samples were taken for chemical analysis.

The soil samples prepared above were digested with HNO_3 and H_2O_2 by using the USEPA method 3050B [25] for analyzing the total concentrations of cadmium (Cd), lead (Pb), chromium (Cr), copper (Cu) and zinc (Zn). These analyses were done by flame atomic absorption spectrometer (ANA82, Tokyo Photo Electric Co., Ltd.).

To determine arsenic (As) concentration, a hydride generation atomic absorption spectrometer (280 FS AA, Agilent) was used and soil samples were digested by HNO_3, HCl, $HClO_4$, H_2SO_4 and HF [26].

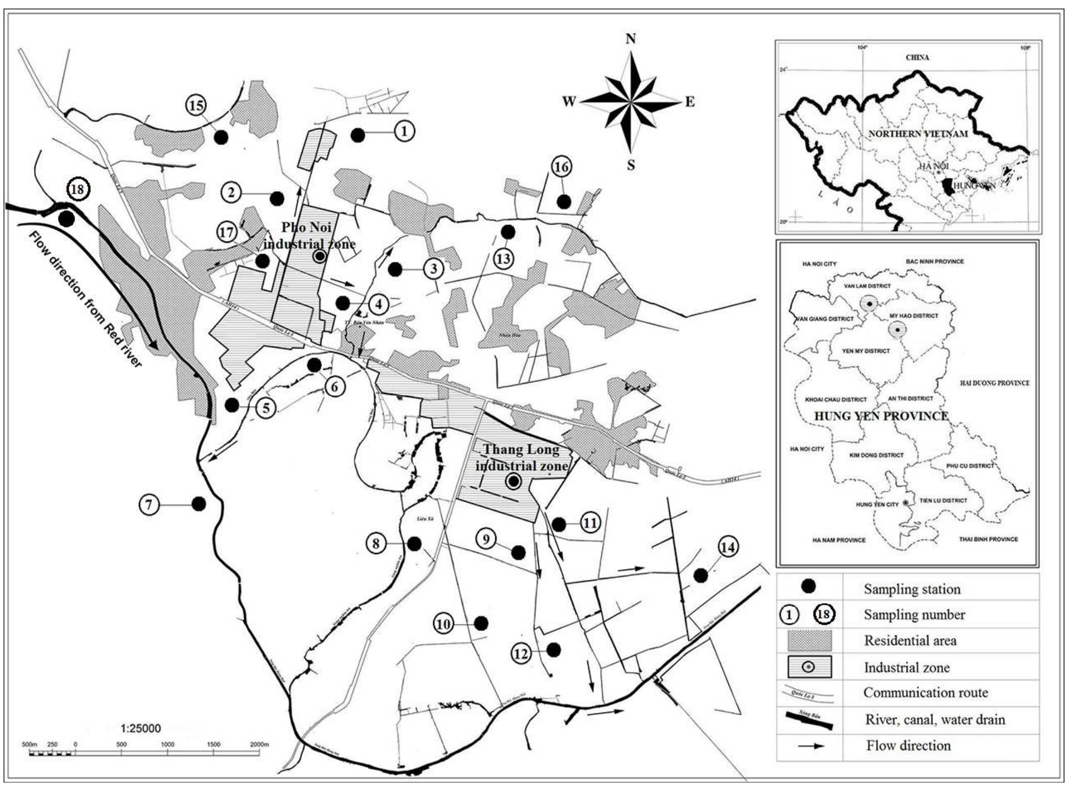

Fig. 1. Study area and soil sample sites in industrial zones in Hung Yen Province, Northern Vietnam

2.3 Data Analysis

A one-way repeated measures analysis of variance (ANOVA) was performed to identify the effect of sampling location on the trace metal concentrations, and Welch's t-test was performed to detect the difference in concentrations between the two industrial park areas. Correlation analysis was performed to detect significant correlation between the trace metal concentrations. All statistical analyses were done through Microsoft Excel.

Furthermore, a Smirnov–Grubbs' outlier test was performed to detect the extreme value in the trace metal concentrations. The extreme value may occur due to possible errors in sampling, measurement and/or data recording. The detected extreme values were excluded from the subsequent analysis. To perform the test, SPSS software was used.

3. RESULTS

Fig. 1 shows the location map of the study district. Irrigation canals are located from east to southeast, covering the district. All 18 points for sampling are indicated in Fig. 1. The total target area was 48.5 km^2.

Fig. 2 shows the trace metal concentrations of Cd, Pb, Cr, Cu, Zn and As at each point. The horizontal axis shows the distance from the irrigation water intake to each point along the irrigation canal. The distance for the canal to reach a point from the irrigation water intake was not limited to one (Fig. 1); however, the canal with the shortest distance was adopted. The points around 9–15 km and 18–27 km in the distance belonged to Pho Noi and Thang Long industrial park areas, respectively. Based on Fig. 2, the Cd, As, Cu, and Pb concentrations were less than 100 $\mu g.g^{-1}$ at all points, while Zn and Cr concentrations mostly exceeded 100 $\mu g.g^{-1}$. At point P1, where the distance from the intake was 0, all concentrations were lower than 111 $\mu g.g^{-1}$, and at point P18, where the distance was about 27 km, the concentrations were as low as those at P1. The Cr and Zn concentrations were high with peaks (peak concentrations were higher than 300 and 240 $\mu g.g^{-1}$, respectively) at a distance around 12 and 24 km, respectively.

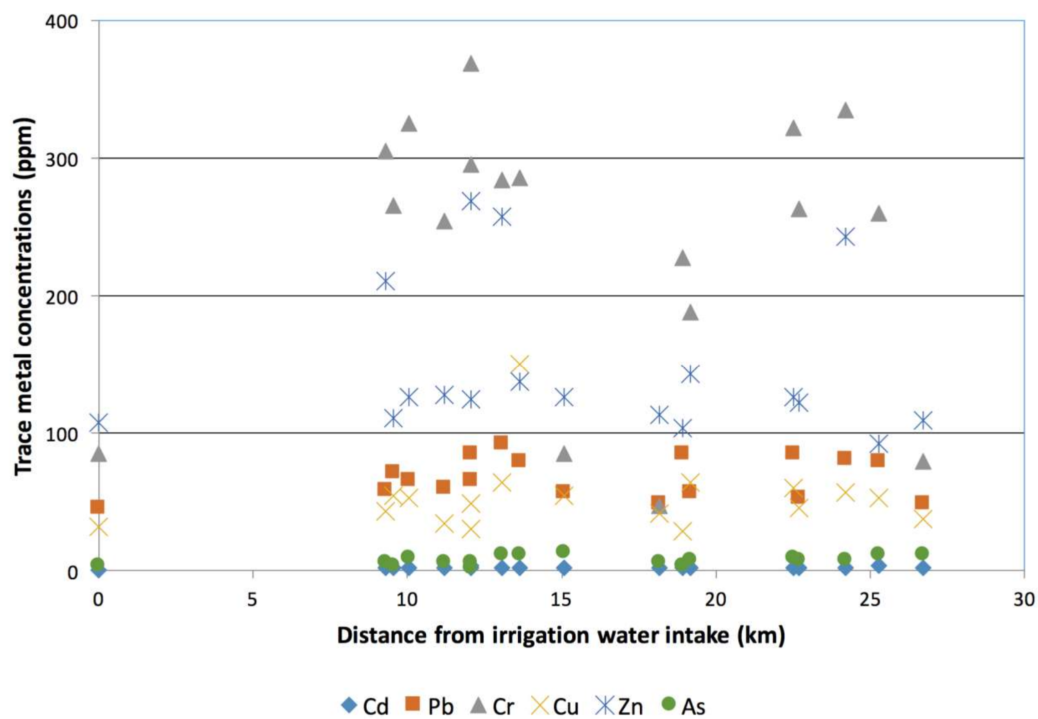

Fig. 2. Trace metal concentrations of Cd, Pb, Cr, Cu, Zn and As in relation to the distance from irrigation water intake

Table 1 shows the trace metal concentrations at respective points and its belonging area, descriptive statistics for the concentrations and some other items.

Table 1 shows that the concentrations of Cd, Pb, Cr, Cu, Zn, and As (µg.g^{-1}) ranged from 1.3–3.5, 45.1–93.2, 46.8–369.0, 29.1–151.3, 93.4–268.3, and 2.3–12.7, respectively. Based on the means, the magnitude of the concentrations was in the order of Cr > Zn > Pb > Cu > Cd > As.

Furthermore, Table 1 shows the permissible level for agricultural soils based on the Vietnamese standard [27], as well as the number of sampling points that the permissible level for each trace metal. In Table 1, every kind of trace metal exceeded the permissible level; arsenic (As) concentration exceeded the level (12 µg.g^{-1}) at one point (sample P3), and the Cd, Cr, and Cu concentrations were exceeded at 10 to13 points out of 18. The maximum exceeding rate of all concentrations was a little higher than 3 times the permissible level occurred in the Cu concentration.

Table 2 shows the summary of the one way repeated measures ANOVA for the effect of sampling points (sampling locations) on the trace metal concentrations. The p value in Table 2 ($p < 0.05$) shows that the effect of sampling locations was significant, i.e., the trace metal concentrations were different between the sampling locations.

According to Table 1, the concentrations were highest at points 1 and 6, and Cd, Cr, and Zn concentrations were highest in P1. Pb had the highest concentration, and Cu, Zn, and As were the second highest in concentration at P6. Concentrations were generally low in P18. More precisely, Cd and Pb were the lowest, and Cr, Cu, Zn and As were the third lowest in concentrations at P18.

Table 1. Trace metal concentrations at respective sampling points, their descriptive statistics and some other items

Sampling points, belonging area, descriptive statistics and other items		Trace metal concentrations (ppm)					
		Cd	**Pb**	**Cr**	**Cu**	**Zn**	**As**
Irrigation water intake	**P1**	**1.3**	**45.1**	**85.2**	**32.4**	**108.1**	**4.2**
Pho Noi area	P2	1.9	58.2	305.4	43.0	211.6	6.3
	P3	2.0	71.3	264.8	55.8	110.9	4.8
	P4	2.8	65.8	325.2	52.8	126.2	9.6
	P5	2.3	61.4	254.3	34.1	128.3	5.8
	P6	1.6	65.8	296.1	50.2	125.0	5.3
	P7	3.5	84.7	369.0	31.1	268.3	2.3
	P8	2.3	93.2	283.7	63.8	258.5	11.8
	P9	2.6	80.0	285.3	151.3	137.6	11.5
	P10	2.2	57.3	85.2	55.6	126.7	12.7
	P11	2.0	50.1	46.8	42.6	113.8	6.3
	P12	2.2	57.6	188.2	63.7	143.3	7.0
Thang Long area	P13	2.3	84.8	227.7	29.1	104.5	3.5
	P14	2.5	84.3	321.9	60.3	125.8	9.3
	P15	1.7	53.7	264.0	45.4	122.5	7.3
	P16	3.1	81.3	334.7	55.9	242.9	7.6
	P17	3.3	79.6	260.0	52.8	93.4	11.4
	P18	1.8	49.2	79.9	38.5	110.2	11.3
Max		3.5	93.2	369.0	151.3	268.3	12.7
Min		1.3	45.1	46.8	29.1	93.4	2.3
Mean		2.3	68.0	237.6	53.2	147.6	7.7
Coefficient of variation		0.25	0.21	0.40	0.49	0.37	0.40
Permissible level for agricultural soils		2	70	200	50	200	12
Number of sampling points exceeded the permissible level		12	8	13	10	4	1

Table 2. Summary of the one way repeated measures ANOVA for the effect of sampling points on trace metal concentrations

Source of variance	**Sum of squares**	**Df**	**Mean square**	**F**	**p value**
Trace metals	743,991	5	148,798		
Sampling points	67,135	17	3,949	1.99	0.02
Trace metals×sampling points	168,707	85	1,985		
Total	979,833	107			

The Smirnov–Grubbs' outlier test was performed separately. As a result, the extreme value (extremely high value) was detected in the Zn concentration at P1, P5, P6, and P11 and in the Cu concentration at P4. The extreme value was not detected in Cd, Pb, Cr, and As concentrations at any point.

Table 3 shows the results of Welch's t-test for the concentrations between the two industrial park areas. According to Table 3, p-values ($p > 0.05$) did not show significant differences between the two industrial park areas for the respective concentrations. This result shows that the concentrations were not different between the two park areas.

Table 4 shows the correlation coefficients between the concentrations of Cd, Pb, Cr, Cu, Zn, and As. According to Table 4, significant correlations were found between Pb, Cd, and Cr at the 1% significant level, and between Cr and Zn, and Cu and As at the 5% significant level, respectively.

4. DISCUSSION

4.1 Source of the Trace Metals

Most of the trace metal concentrations were higher in the central part of the industrial park areas than in the irrigation water intake (Table 1, Fig. 2). High concentrations were observed particularly at points 1, 2, 4, 9, 11, and 12 in the central part (Table 1). These points were located near factories (Fig. 1); therefore, the high concentrations were thought to be caused by the entry of the factory-discharged wastewater into the paddy field. As mentioned previously, the district is prone to flooding, and all wastewater from the industrial zone were directly discharged to the irrigation system. Therefore, the entry of heavy metal in soils at these points was probably caused by the flooding of a mix of wastewater and irrigation water and the long-term application this mixture as the source of the irrigation water. The concentration was low at the points away from factories (Fig. 1), indicating that the soil contamination was restricted to near the factory areas.

The extremely high Zn concentration was observed at points 1, 5, 6, and 11 based on the outlier test, indicating that the factories discharging Zn-containing wastewater were located near these points. The extremely high Cu concentration was observed at point 4 by the test, indicating that the factories discharging Cu-containing wastewater were located around this point.

Table 3. Results of Welch's t-test for Cd, Pb, Cr, Cu, Zn, and As concentrations between the two industrial parks

Trace metals	Industrial Park	t	d.f.	p value
Cd	Pho Noi	0.52	8.3	0.61
	Thang Long			
Pb	Pho Noi	0.56	8.74	0.59
	Thang Long			
Cr	Pho Noi	0.05	11.21	0.96
	Thang Long			
Cu	Pho Noi	1.06	13.74	0.31
	Thang Long			
Zn	Pho Noi	0.91	10.93	0.38
	Thang Long			
As	Pho Noi	0.51	11.56	0.62
	Thang Long			

Table 4. Correlation coefficients between the trace metal concentrations of Cd, Pb, Cr, Cu, Zn, and As

	Pb	Cr	Cu	Zn	As
Cd	0.69**	0.55*	0.16	0.40	0.15
Pb		0.70**	0.29	0.47	0.09
Cr			0.19	0.51*	-0.14
Cu				0.01	0.51*
Zn					-0.09

** and **: significant at 5 and 1 % levels*

Based on the correlation coefficients (Table 4), there were three groups of trace metals, i.e., a group of Cd, Pb, and Cr, a group of Cr and Zn, and a group of Cu and As, based on the significant correlations. The classification of the groups shows that there were several groups of wastewater containing trace metals. In particular, high concentrations of Cr and Zn with significant correlation show that the trace metals may be supplied from the same source of wastewater. Cr and Zn are thought to be discharged from the metal plating processes in a factory, which may be one of the mechanical factories mentioned in the study area, and is present in both industrial parks.

4.2 Difference in the Trace Metal Concentrations between the Two Industrial Park Areas

The trace metal concentrations were in the order of Cr > Zn > Pb > Cu > As > Cd for the respective industrial park areas, based on the average concentrations of the respective areas separately calculated from Table 2. Furthermore, no significant differences were observed in the respective concentrations between the two industrial park areas (Table 3).

Based on these characteristics, the factory composition and foundation year most likely did not affect our results. Here, the absence of any effects from factory composition may have been caused by the concentrations becoming similar with each other after the mixing of wastewaters in irrigation channels in the respective industrial park areas. The absence of effects due to the foundation year (the passage of time), where there is a difference of six years between foundation, may be due to the absorption of trace metals by crops, and/or loss of trace metals by soil washing from flooding. This could decrease the concentrations so that they reached similar levels between the two areas. Usually, the soil trace metal concentrations are considered to increase over time, which has previously been mentioned as a long-term effect of irrigation in trace metal contamination.

4.3 Usability of the Soils for Agricultural Purposes

The Cd, Cu, Pb, Zn, and As concentrations exceeded the permissible levels for agricultural soils in Vietnam at 1–13 points out of 18 (Table 1). Although these metal concentrations of some soil samples did not exceed these levels, but many concentrations were close those presented in Table 1. Despite a lack of permissible levels for Cr soil concentration in Vietnam, Cr concentrations exceeded 200 $\mu g.g^{-1}$ which was the highest permissible range determined by Kabata-Pendias [28] at many points. The soils in this district, contaminated by all trace metals, are therefore inadequate for agricultural soils, and soil decontamination measures are necessary.

5. CONCLUSIONS

The following conclusions were drawn from the study. The magnitude of the trace metal concentrations in the soil samples was in the order of Cr > Zn > Pb > Cu > As > Cd for the study district. Concentrations were highest at the center of the industrial park areas, where there are many factories compared to those in the irrigation water intake, indicating that the trace metals originated from factories within the industrial parks with wastewater. Even inside the district, concentrations were low at points away from the factories, indicating that the contamination was restricted to factory areas.

No significant differences were observed between the two industrial park areas in the concentrations despite differences in foundation year and factory composition between the two industrial park areas.

The trace metal concentrations of the soils exceeded or were close to the permissible levels for agricultural purposes, thus the soils are not adequate for agriculture and some remediation measures, such as soil decontamination, are necessary.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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