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HEAVY METAL ACCUMULATION IN THE AGRICULTURAL SOIL AROUND IRON-RECYCLING CHAU KHE VILLAGE IN TU SON TOWN, BAC NINH PROVINCE, VIETNAM

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ABSTRACT

The present study targeted Chau Khe craft village of Tu Son town, Bac Ninh province, Vietnam, where the recycled-steel craft has been performed widely for a long time, to evaluate accumulation of heavy metals such as Cd, Cu, Pb and Zn in the agricultural soil surrounding the iron-recycling area of Chau Khe. Total 12 soil samples was collected at the agricultural field around Chau Khe village in 2010 and 2015, followed by analyses of the selected physico-chemical properties and the concentration of heavy metals in the soil. Results showed that the concentations of total heavy metals in 2015 were all at a high level in the agricultural soil, with averaged values of 0.47 mg/kg for Cd, 41.72 mg/kg for Cu, 34.77 mg/kg for Pb and 320.19 mg/kg for Zn, and exceeded by 1.5 times the threshold concentrations allowed by QCVN03:2015/BTNMT - Vietnam National Technical Regulation on permissible limits of heavy metals in the soil. The total concentration increased for Cu and Zn, whereas decreased for Pb and Cd with time. The concentration of a soluble form increased only for Zn and decreased for Pb, Cu and Cd with time. Correlation analysis indicated that the physico-chemical properties of the agricultural soil were correlated more closely with the heavy metal concentration of a soluble form than with the total heavy metal concentration.

Keywords: Chau Khe, agriculture soil, heavy metals, village, recycled metal.

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1. INTRODUCTION

Research on the heavy metal accumulation in the agricultural soil has been conducted with great interest by many scientists all over the world. Heavy metal pollution in the agricultural soil can come from various sources such as natural origin, chemical fertilizers (Alloway, 1990; Nriagu, 1988), industry, mining (Erick Millstone, 1997) and transportation (Virginiaw - Maclaren, 1994). In Vietnam, the heavy metal concentration of agricultural soil does not reach an alarming level but is in a increasing trend (Ngo Thi Bao Minh et al., 2015). According to the report on the environmental status of Vietnam in 2015, several areas of agricultural land in localities such as Lam Dong, Ha Noi and Lang Son have the As and Cu concentrations exceeding the allowable limit (Ministry of Natural Resources and Environment of Vietnam, 2016). The heavy metal pollution in Vietnamese soil is mainly due to the impact of application of chemical fertilizers, industrial discharge and house-level metal recycling (Ngo Thi Bao Minh et al., 2015; Ministry of Natural Resources and Environment of Vietnam, 2016; Ho Thi Lam Tra, 2005). Accumulation of heavy metals in the agriculture soil has potentially an adverse impact on the agricultural production and human health. Therefore, proper monitoring of the accumulation of heavy metals in the agricultural environment plays a significant role on the protection of agricultural environmentat and prevention of health damage of local people from environmental pollution. Our study focused on a recycling village of Chau Khe in Bac Ninh province, where iron recycling in a house-level has been performed intensively for many years, in order to conduct (i) monitoring the level of heavy metal accumulation in the agricultural soil and (ii) making recommendations for agricultural environment conservation of the targeted area to the local government and people.

2. RESEARCH METHODS

2.1. Study area

Chau Khe craft village in Tu Son town, Bac Ninh province is located in the central region of the Red River delta of Vietnam (Figure 1) with total natural area of 495.86 ha and population of 15,178 in 2014 (Data statistic of Bac Ninh provice of Vietnam, 2015). The Red River delta is under the tropical monsoon climate with two seasons of the rainy season from April to October and the dry season from November to March. In Chau Khe village, the annual mean rainfall is about 1400-1600 mm concetrating on three months of June through August the annual mean temperature is 23.3^{o}C, and the annual mean air humidity is 88% (Department of Natural Resources and Environment in Bac Ninh of Vietnam, 2016).

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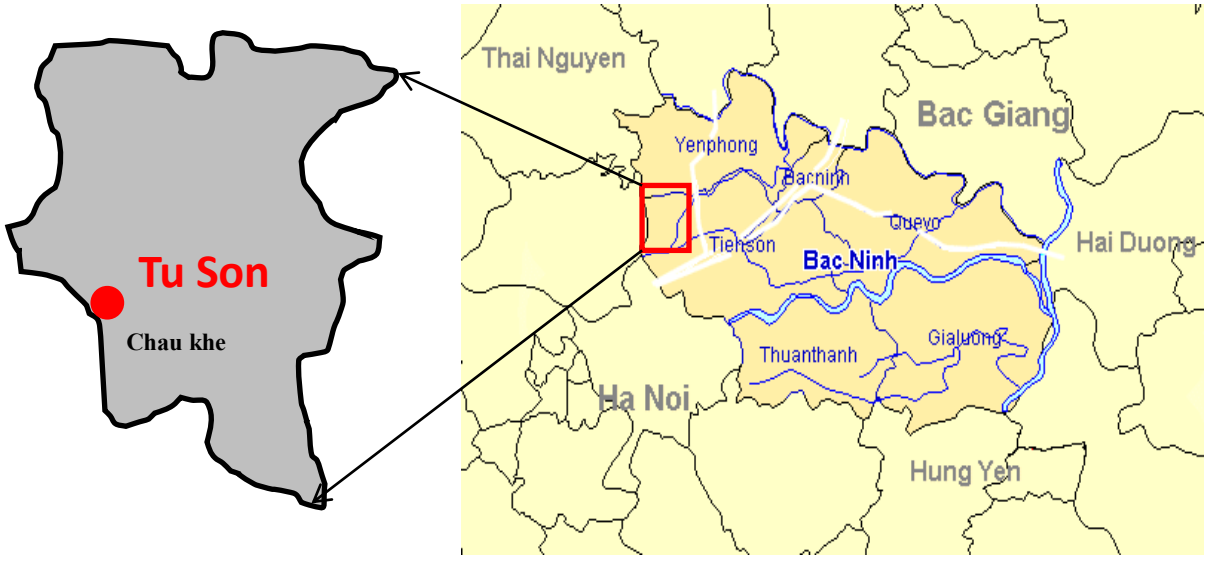

Figure 1: Location of Chau Khe village on the map of Tu Son town, Bac Ninh province of Vietnam

2.2. Secondary data collection

The secondary data concerning the statistics on iron recycling and production activities of Chau Khe village were collected from People's Committee of Chau Khe ward and Department of Natural Resources and Environment of Bac Ninh province of Vietnam.

2.3. Soil sampling

To conduct the present study, soil samples were taken at 12 sites of the agricultural field around the Chau Khe village in 2010 and 2015. Soil samples were collected from the topsoil, according to the soil sampling procedure of the ISO 10381 - 1:2002. Soil samples were then dried in the air, ground and sieved through a 2-mm sieve for further analysis of the soil quality indicators.

2.4. Soil analyses

The total concentration of Cd, Cu, Pb and Zn was measured after dissolution with concentrated HCl and HNO_3 having a ratio of 3:1 (ISO 11466:1995). The concentration of soluble forms of Cd, Cu, Pb and Zn was measured after extraction with 0.1 M hydrochloric acid under the acid:soil ratio of 10:1. The heavy metal concentration was determined by using an atomic absorption spectrophotometer (AAS) at a wavelength of 228.8 nm for Cd, 324.8 nm for Cu, 217.0 nm for Pb, and 213.9 nm for Zn (EPA Method 7000B). The analysis was conducted in

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duplicate and the standard deviation of the measured values did not exceed 5%. The heavy metal concentrations thus obtained were compared with QCVN03:2015/BTNMT – Vietnam National Technical Regulation on permissible limits of heavy metals in the soil.

Physico-chemical analysis of the soils samples was as follows: pH was measured by the pH meter glass electrode with a soil: H_2O ratio of 1:5 (ISO 10390: 2005); organic matter content (OM) was determined by the Walkley-Black method (Walkley and Black, 1934); and cation exchange capacity (CEC) was determined according to the ISO 11260:1994.

2.5. Statistical analysis

The description, correlation analysis and testing of analytical data were performed by Stata 2010 software.

3. RESEARCH RESULTS

3.1. Statistical data on iron recycling and production activities in Chau Khe village

Chau Khe village has a long history of iron recycling. Some statistical data on iron recycling and production activities in Chau Khe village (Department of Natural Resources and Environment in Bac Ninh province of Vietnam, 2016) are shown in Table 1. According to the statistics shown in Table 1, Chau Khe village had more than 1,700 households engaging in iron recycling in 2010. Due to fluctuation of the national economy and the pressure of the environmental conservation, however, the corresponding number of households in Chau Khe village decreased sharply to 562 in 2015. In accordance with the sharp decrease in the number of households engaging in iron recycling, indices (or items) such as the amount of waste discharged from iron recycling and the weight of solid waste (coal slag and dross) generating from production activities decreased rapidly in 2015. It suggests improvement of agricultural environment around Chau Khe village in 2015.

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Table 1: Some statistical data on iron recycling and production activities in Chau Khe village of Tu Son town, Bac Ninh province of Vietnam

Indices	Unit	2010	2015	Increase/Decrease
Area subjected to production activities	ha	6.51	32.11	+ 25.60
Number of households engaging in iron recycling	number	1,767	562	-1,205
Amount of waste discharged from iron recycling	ton	350,000	150,000	-200,000
Number of households whose income from recycled iron is over 20 billion VND/year	number	472	128	-344
Weight of solid waste (coal slag and dross) generating from production activities.	ton/household/year	50	14	-36
Volume of water consumed	m^3/household/year	2,700	800	- 1,900
Weight of gas and dust emitting	ton/household/year	267	152	- 115

Department of Natural Resources and Environment in Bac Ninh province of Vietnam, 2016

3.2. Selected physico-chemical properties of the agricultural soil

Selected physical and chemical properties of the agricultural soil around Chau Khe village in 2010 and 2015 are shown in Table 2. Soil pH was 4.50 in average, ranging from 3.89 to 6.21, in 2015, which was low compared to the average value of 5.07 in 2010. Cation exchange capacity (CEC) ranged from 7.83 to 20.84 meg/100-g soil (11.09 meg/100-g soil in average) in 2015. Exchangeable cations were at a low or moderate level under the FAO's rating scale ranging from 2.48 to 14.58 meg/100-g soil for $Ca2^+$; from 0.74 to 1.81 meg/100-g soil for Mg^{2+} from 0.20 to 3.33 meg/100-g soil for K^+; and from 0.19 to 0.83 meg/100-g soil for Na^+. In general, exchangeable cations were higher for 2015 than for 2010.

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Table 2: Selected physico-chemical properties of the agriculture soil around Chau Khe village in Tu Son town, Bac Ninh province of Vietnam

Value	pH		OM (%)		CEC (meg/100-g soil)		Exchangeable cations (meq/100-g soil)							
							Ca^{2+}		Mg^{2+}		K^{+}		Na^{+}	
	2010	**2015**	**2010**	**2015**	**2010**	**2015**	**2010**	**2015**	**2010**	**2015**	**2010**	**2015**	**2010**	**2015**
Min	4.32	3.89	2.40	2.16	5.86	7.83	0.07	2.48	0.01	0.74	0.12	0.20	0.12	0.19
Max	5.82	6.21	4.40	6.14	13.66	20.84	0.26	14.58	0.04	1.81	0.62	3.33	1.00	0.83
Ave	5.07	4.50	3.34	3.12	9.93	11.09	0.16	5.76	0.03	1.16	0.25	0.56	0.30	0.39
SD	0.50	0.80	0.54	1.02	2.23	3.55	0.06	3.27	0.01	0.30	0.14	0.88	0.25	0.16

Note: n = 12 (number of soil sample), Min: Minimum, Max: Maximum, Ave: Average, SD: standard Deviation, OM: Organic Matter, CEC: Cation exchange capacity.

3.3. Concentrations of heavy metals in the agricultural soil

Table 3 shows the analytical results of the total (TS) and soluble (DT) concentrations of heavy metals (Cu, Pb, Zn and Cd) in the agricultural soil around Chau Khe village. According to the data shown in Table 3, the average total concentrations in 2010 and 2015 were 40.52 and 41.72 mg/kg for Cu, 47.96 and 34.77 mg/kg for Pb, 195.39 and 320.19 mg/kg for Zn, and 0.61 and 0.47 mg/kg for Cd. These results were similar to the total heavy metal analysis done in 2014 by Institute of Agricultural Environment at the same location (Ministry of Natural Resources and Environment of Vietnam, 2016). Meanwhile, the average soluble concnetrations were 16.29 mg/kg in 2010 and 13.46 mg/kg in 2015 for Cu, 17.02 mg/kg in 2010 and 9.32 mg/kg in 2015 for Pb, 70.71 mg/kg in 2010 and 196.91 mg/kg in 2015, and 0.20 mg/kg in 2010 and 0.06 mg/kg in 2015 for Cd.

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Table 3: The concentration of heavy metals in the agricultural soil around Chau Khe village in Tu Son town, Bac Ninh province of Vietnam

Year	Value	Cu			Pb			Zn			Cd		
		Total	Soluble	Ratio*	Total	Soluble	Ratio*	Total	Soluble	Ratio*	Total	Soluble	Ratio*
2010	Min	33.19	13.10	39.47	36.24	13.10	36.15	121.56	23.78	19.56	0.47	0.14	29.79
	Max	53.78	22.32	41.50	61.55	22.32	36.26	457.51	223.51	48.85	0.86	0.24	27.91
	Ave	**40.52**	**16.29**	**40.19**	**47.96**	**17.02**	**35.48**	**195.39**	**70.71**	**36.19**	**0.61**	**0.20**	**33.29**
	SD	6.32	2.72	43.05	6.73	2.91	43.21	93.77	53.03	56.55	0.13	0.04	26.87
2015	Min	29.06	8.01	27.56	26.78	5.87	21.92	112.93	35.84	31.74	0.30	0.04	14.00
	Max	72.95	17.87	24.50	46.48	14.89	32.04	690.50	513.63	74.39	0.63	0.09	13.97
	Ave	**41.72**	**13.46**	**32.27**	**34.77**	**9.32**	**26.80**	**320.19**	**196.91**	**61.50**	**0.47**	**0.06**	**13.18**
	SD	11.06	2.64	23.91	6.35	2.63	41.49	202.88	157.59	77.68	0.12	0.02	12.73

Notice: n = 12; (*) = % ratio of the soluble to total concentrations; Min: Minimum; Max: Maximum; Ave: Average; SD: standard Deviation; OM: Organic Matter; CEC: Cation exchange capacity.

In 2015, it is remarkable that the percent ratio of the soluble to total concentrations still remained at a relatively high level. The soluble concentration was equivalent to 32.3, 26.8, 13.2 and even 61.5% of the total concentration for Cu, Pb, Cd and Zn, respectively. The soluble and total concentrations of Cu, Pb, Zn and Cd at each sampling site in the agricultural soil ~~of~~ around Chau Khe village in 2015 are presented in Figure 2. In most sampling sites, the soluble concentrations of Cu and Zn were in a relatively high level in comparison with their total concentrations. This relatively high proportion of the soluble heavy metals might have the ability to create pollutionin in a large scale and affect the quality of agricultural products (Chu Anh Dao et al., 2012).

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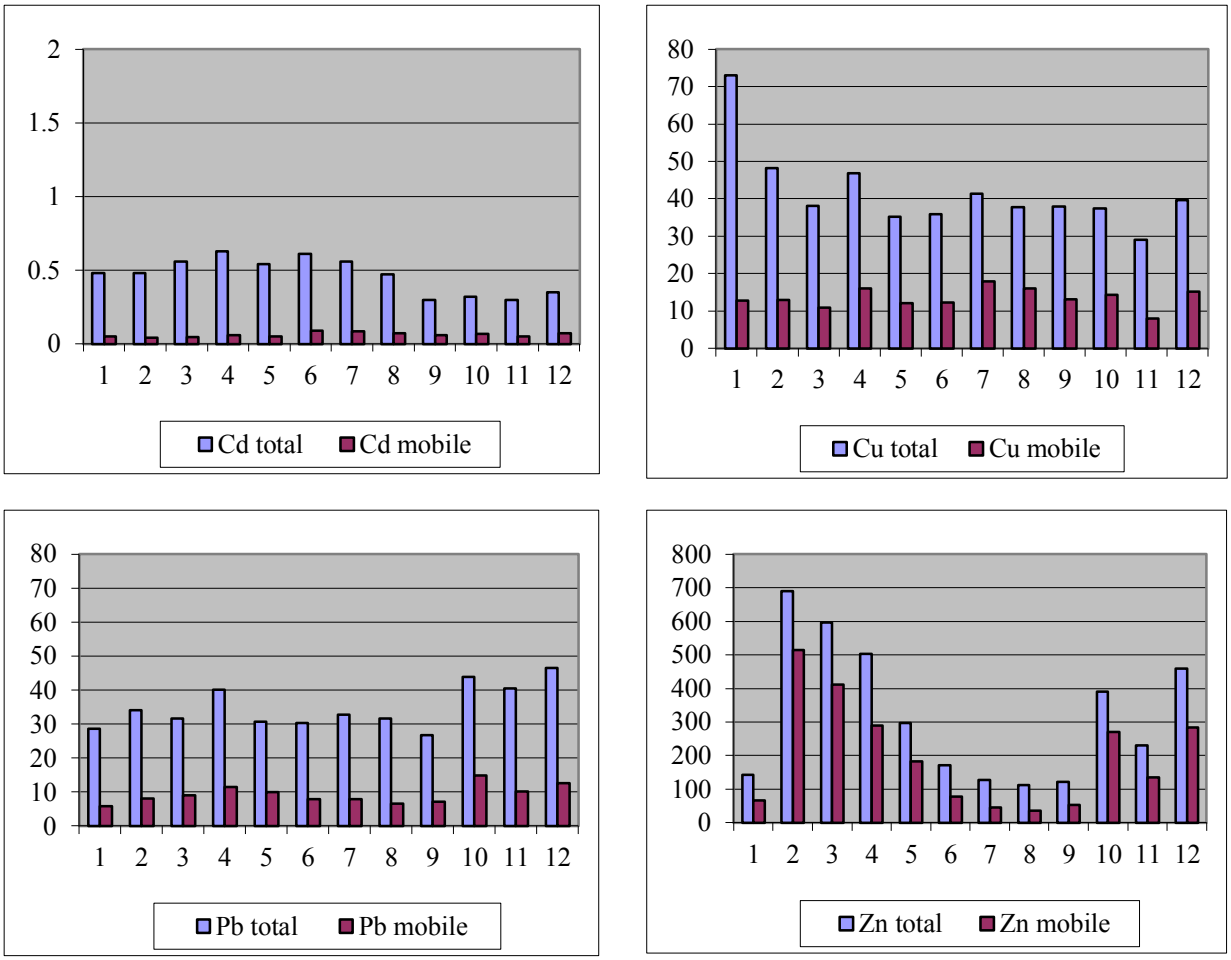

Figure 2: Soluble and total concnetrations of heavy metals (Cd, Cu, Pb, and Zn) at each sampling site in the agricultural soil around Chau Khe village in Tu Son town, Bac Ninh province of Vietnam in 2015

4. DISCUSSION

4.1. Accumulation of heavy metals in the agricultural soil

Results in Table 3 indicated that the heavy metal concentrations in the agricultural soil around Chau Khe village were higher than those generally reported to the agricultural soil of Vietnam. It suggests that metal recycling activities of Chau Khe village have a major influence on the accumulation of heavy metals in the agricultural soil around the village. This tendency is in a complete correlation with other studies on the heavy metal accumulation in the agricultural soil around the villages of metal recycling, such as bronze casting village in Dai Dong commune,

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Hung Yen province (Le Duc and Nguyen Ngoc Minh, 2001), lead recycling village in Chi Dao commune, Hung Yen province (Dang Thi An and Tran Quang Tien, 2008; Ho Thi Lam Tra, 2005), aluminum and bronze casting village in Van Mon commune, Bac Ninh province (Pham Quang Ha, 2003), in Vietnam.

In Chau Khe village, there was observed an increase in the total concentrations of Cu and Zn with time (The total concentration of Cu is hardly different between 2010 and 2015), whereas the total concentration of Pb showed a decreasing trend. Regarding the soluble heavy metal concentration, the concentration of Zn increased with time, whereas the concentrations of Cu, Pb and Cd tended to decrease with time. To examine the statistical difference of the average values of soluble and total heavy metal concentrations between 2010 and 2015, the two-dimension t-test was conducted at the significant level of 95%. The analytical results indicated that only the total concentration of Pb had the significantly ($P = 0.00001$) different average values between 2010 and 2015 with showing a decreasing trend with time. The average of the total concentrations of other three heavy metals was not significantly different between 2010 and 2015. In contrast, the average values of the soluble concentration were significantly different between 2010 and 2015 for all heavy metals investigated at the 95% significant level, and the average soluble concentration decreased for Cu ($P = 0.0284$), Pb ($P = 0.00001$) and Cd ($P = 0.0003$) but increased for Zn ($P = 0.0691$) with time. The temporal increase of the soluble Zn concentration is very serious, because Zn can be absorbed by plants and accumulated within the plant body to increase the contamination risk of agricultural products and impact on human health.

The temporal variation of the heavy metal concentrations observed in the agricultural soil around Chau Khe village is quite complex. However, it can be seen that the total heavy metal concentrations (Cu, Pb, Zn and Cd) in the agricultural soil around Chau Khe village are much higher than those reported to cultivated agricultural soils in Vietnam. Although the production scale of Chau Khe village has been decreased by more than 50% and solid waste generating also decreased by more than 40% (Table 1), the concentrations of heavy metal such as Zn tended to increase. This confirms that a large amount of Zn has been accumulated in the agricultural soil over time.

4.2. Quality assessment of agricultural soil according to QCVN03

According to QCVN03:2015/BTNMT - Vietnam National Technical Regulation on permissible limits of heavy metals in the soil, the allowed threshold for the total concentrations of heavy metals in the agricultural soil is 100, 70, 200 and 1.5 mg/kg for Cu, Pb, Zn and Cd, respectively. Comparison of these values with the present analytical results indicates that the average total concentrations of Cu, Pb and Cd (both in 2010 and 2015) remain below the prescribed threshold.

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However, the average total concentration of Zn increased and was much higher for 2015 than for 2010 to exceed the allowable limits in 2015. Thus, it can be mentioned that the agricultural soil around Chau Khe village was contaminated by Zn in 2015 (Figure 3). Unfortunately, both soluble and total concentrations of tended to increase with time. The agricultural soil polluted by Zn will increase the risk of Zn accumulation in crops and have a negative impact on human health. According to the report of Vietnam's Ministry of Health, the Zn concentration in a range of 5-10 ppm in food is safe for the human health. However, if the concentration of Zn in food becomes too high, it can cause acute poisoning in humans with symptoms such as vomiting, diarrhea, cold sweat and pulse softly, and even cause death.

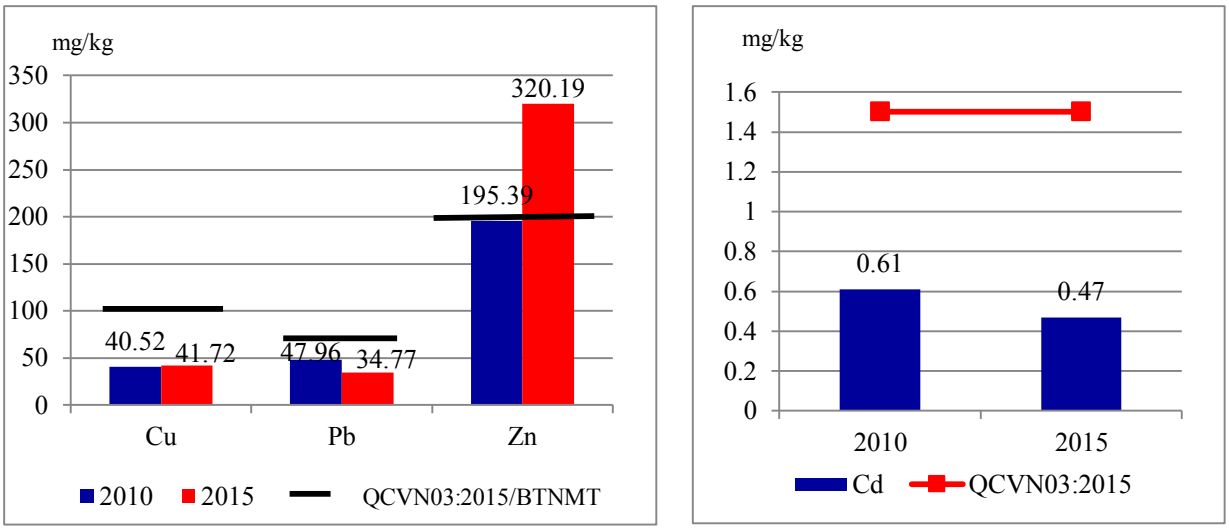

Figure 3: Comparison of the average total concentrations of heavy metals in the agricultural soil around Chau Khe village with QCVN03:2015/BTNMT

Notice: QCVN03:2015/BTNMT- Vietnam National technical regulation on the allowable limits of heavy metals in the agricultural soils

The total concentrations of other heavy metals such as Cu, Pb and Cd remained in the allowable limits of QCVN03/2015/BTNMT. However, Cu and Pb concentrations were at a relatively high level, closing to the threshold. Therefore, it is essential to pay attention to impose warnings on time.

4.3. Relationship between soil properties and heavy metals concentrations

Many scientific reports over the world have shown that physical and chemical properties of soil are related to the composition and form of heavy metals in a soil (Loretta Y.Li and Raymond, 2000). According to Plant and Raiswell (1983), many heavy metals are mobilized under highly

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acidic (low pH) and oxidizing conditions while immobilized under alkaline and reducing conditions Cherry et al. (2014) reported that mobilization of Zn^{2+} in soil is affected by the contents of iron and aluminum oxides, while that of Cu^{2+} is affected by soil composition and of Pb^{2+} by the contents of aluminum and iron oxides and organic matter in soil.

We examined correlation of heavy metals concentrations with some physical and chemical properties of the agticultural soil around Chau Khe village. Correlation coefficients are shown in Table 4. The total concentrations of almost all heavy metals did not have a significant correlation with soil physical and chemical properties, except the total concentraion of Zn which was significantly correlated with the contents of exchangeable Mg^{2+} (r = -0.41) and Na^{+} (r = 0.45) and CEC (r = -0.46) at the 5% significant level and with pH (r = -0.56) and the content of exchangeable Ca^{2+} (r = -0.53) at the 1% significant level. Almost all correlations were negative, and only the correlation ~~to~~ with the content of exchangeable Na^{+} was positive. In contrast, the correlation between soil physical and chemical properties and the soluble heavy metal concentrations was quite clear, namely the negative Cu_s significantly correlated with OM (r = -0.64) and positively correlated with Mg^{2+} (r = 0.49); Zn_s negatively correlated significantly with both pH (r = -0.56) and Ca^{2+} content (r = -0.56); and negatively correlated with the concentration of Mg^{2+} weak (r = -0.45), CEC (r = -0.49) and positive correlated with Na^{+}(r = 0.42); Regarding the content of Cd_{dt}, its content correlated significantly with pH (r = 0.82), content of Ca^{2+} (r = 0.78), Mg^{2+} (r = 0.52), K^{+} (r = 0.56) and CEC (r = 0.72) in soil.

Table 4: Correlation coefficients between heavy metals concentrations and physico-chemical properties of the agricultural soil around Chau Khe village in Tu Son town, Bac Ninh province of Vietnam

	pH(KCl)	OM(%)	Ca^{2+}	Mg^{2+}	K^{+}	Na^{+}	CEC
Cu_t	-0.21	-0.38	-0.17	-0.18	-0.18	-0.11	-0.32
Pb_t	-0.36	0.25	-0.33	-0.08	-0.17	0.58*	-0.15
Zn_t	-0.56*	-0.13	-0.53*	-0.41*	-0.25	0.45*	-0.46*
Cd_t	0.32	-0.40*	0.45*	0.21	0.33	-0.25	0.35
Cu_s	0.36	-0.64*	0.21	0.49*	-0.15	0.06	0.001
Pb_s	-0.34	0.16	-0.35	-0.34	-0.13	0.37	-0.22
Zn_s	-0.56*	-0.10	-0.56*	-0.45*	-0.29	0.42*	-0.49*
Cd_s	0.82*	-0.23	0.78*	0.52*	0.56*	-0.08	0.72*

Notice: OM:organic matter; Ca^{2+}: calcium ion; Mg^{2+}: magnesium ion; K^{+}: Kalium ion; Na^{+}: Natrium ion; CEC: Cation exchange capacity; t=total; s:soluble; (*) significantly corelation.

The correlation analysis revealed a close relationship of soil physical and chemical properties with the soluble concentrations of heavy metals. Based on this, we can propose some methods to

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mitigate the adverse impact of heavy metal accumulation in soil through improvement of soil properties. For instance, improving soil properties to convert heavy metals from a soluble form to a insoluble form can limit the heavy metal absorption by plant, thereby reducing risk to human health.

5. CONCLUSIONS & RECOMMENDATIONS

5.1. Conclusions

The heavy metal concentrations in the agricultural soil around Chau Khe village in Tu Son town, Bac Ninh province of Vietnam were in a relatively high level, and the average total and soluble concentrations were 41.72 and 13.46 mg/kg for Cu, 34.77 and 9.32 mg/kg for Pb, 320.19 and 196.91 mg/kg for Zn, and 0.47 and 0.06 mg/kg for Cd. Among the four heavy metals targeted, the total concentration of Zn in 2015 exceeded by 1.5 times the allowed threshold of QCVN03:2015/BTNMT - Vietnam National Technical Regulation on permissible limits of heavy metals in the soil.

The total concentration tended to increase for Cu and Zn while decrease for Pb and Cd with time. Meanwhile, the soluble concentration tended to decrease ~~of~~ for Cu, Pd and Cd while increase for Zn with time. The result of the two dimensionalt-test at the significance level of 95% showed that the temporal variation of the total Cu concentration was significant, while those of Zn, Pb and Cd were not significant. In contrast, the temporal variation of the soluble concentration was significant for all heavy metals.

The analytical result of the correlation of heavy metals concentrations with physico-chemical properties of soil indicated that the total concentration was little related to the soil properties but that the soluble concentrations of almost all heavy metals were correlated with the soil characteristics.

5.2. Recommendations

Soil improvement is essential to reduce accumulation of heavy metals in agricultural soil. It is necessary to pay attention to some key ideas: improving soil properties to convert heavy metals from soluble to insoluble forms to limit absorption by plant. Waste gas and water, and solid waste from Chau Khe village are needed to be handled thoroughly to avoid soil pollution. Making recommendations to farmers and those involved on the current status of Zn pollution in Chau Khe village for timely response measures.

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