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Heavy Metal of Soil in Wastewater – Irrigated Agricultural Soil in a Surrounding Area of the Nhue River, Vietnam

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Waste from industrial sources, serves as sources of water for irrigating farms. The purpose of this study is to identify the impact of waste–water irrigation on the level of heavy metals in the soils. Soil samples were collected from the different locations from upstream to downstream of the Nhue River to evaluate heavy metal pollution. The results showed that the concentrations of all heavy metals in the soil samples in the farmland area were much higher than the background level in that area (1.2–2.6 mg/kg for Cd, 42–60 mg/kg for Cr, 22–62 mg/kg for Cu, 30–86 mg/kg for Pb, 119–245 mg/kg for Zn, and 26–57 mg/kg for Ni), and exceeded the level of Vietnamese standard for agricultural soil for all heavy metals Cd, Cu, Pb and Zn except soil samples at upstream and downstream of the Nhue River.

Key words: heavy metal, Nhue River, soil, wastewater irrigation

INTRODUCTION

Heavy metal contamination of soil resulting from wastewater irrigation is a cause of serious concern due to the potential health impacts of consuming contaminated produce. In suburban areas, the use of industrial or municipal wastewater is common practice in many parts of the world (Feigin *et al.*, 1991), including Vietnam (Nguyen *et al.*, 2007). Access to adequate water for irrigation is a matter of increasing concern in Vietnam. To face the growing demand for irrigation water, non-conventional resources are often used. Importance sources of heavy metals in wastewater are urban and industrial effluents, deterioration of sewage pipe and treatment works, and the wear of household plumbing fixtures. Wastewater irrigation is known to contribute significantly to the heavy metal content of soils (Singh *et al.*, 2004). A number of previous studies from developing countries have reported heavy metal contamination in wastewater and wastewater-irrigated soil (Mapanda *et al.*, 2005; Singh *et al.*, 2004). However, there are very few empirical data from Vietnam for heavy metal contamination of water and its transfers to the soil.

Accumulation of heavy metals in the soil, especially in upper horizons, eventually could cause serious problems, resulting in health risks if they enter the food chain. Heavy metal concentrations in soil are typically

quantified and regulated on the basis of total metal content, regardless of their solubility. However, soils with large amount of colloidal organic and mineral material can sorb and immobilize these metals to greater extent than soils with a low content of these reactive materials.

Recently, increasing industrialization and population growth have led to increasing fluxes of many heavy metals to soils of the To Lich and Kim Nguu river system in Hanoi City, Vietnam (Ho and Egashira, 2000). This has also great relevance to the Hanoi agro–ecosystem, where the wastes produced in urban and industrial areas provide the most likely sources of heavy metal pollution, and the dominant sources of soil heavy metal pollution are sewage irrigation. Water used for irrigation in agricultural soil of Hanoi City is seriously polluted by industrial wastewater (Nguyen *et al.*, 2007). In the long term, irrigation may cause the accumulation of heavy-metals at toxic concentrations in the soil and adversely affect both soil microbiological processes and plant growth.

The present study is based on agricultural soil at the surrounding area of the Nhue River, Vietnam which is estimated as one of the most polluted agricultural soil areas, and where wastewater from the Nhue River has been commonly used for irrigation of peri–urban crops for several decades. Study on the pollution of the Nhue River in Vietnam has a relatively short history and has been conducted by a few authors. Kikuchi *et al.* (2009) discussed heavy metal pollution in the water of the Nhue River by using multivariate analyses. Trinh *et al.* (2007) studied about the impact of waste water from industrial activities. The objective of this study is to assess the effects of polluted irrigation water on the quality of farmland soil in the Nhue River basin.

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MATERIALS AND METHODS

Materials

Forty surface soil samples and surface water samples were collected from 10 locations along the Nhue River, referred as SS1 to SS10 and WS1 to WS10 (Fig. 1). At each location samples were taken in four times: December 2011; March, June and August 2012. The soil samples were air-dried, disaggregated, passed through a 1-mm sieve, and preserved in plastic bottles at room temperature for analysis (Ho *et al.*, 1998).

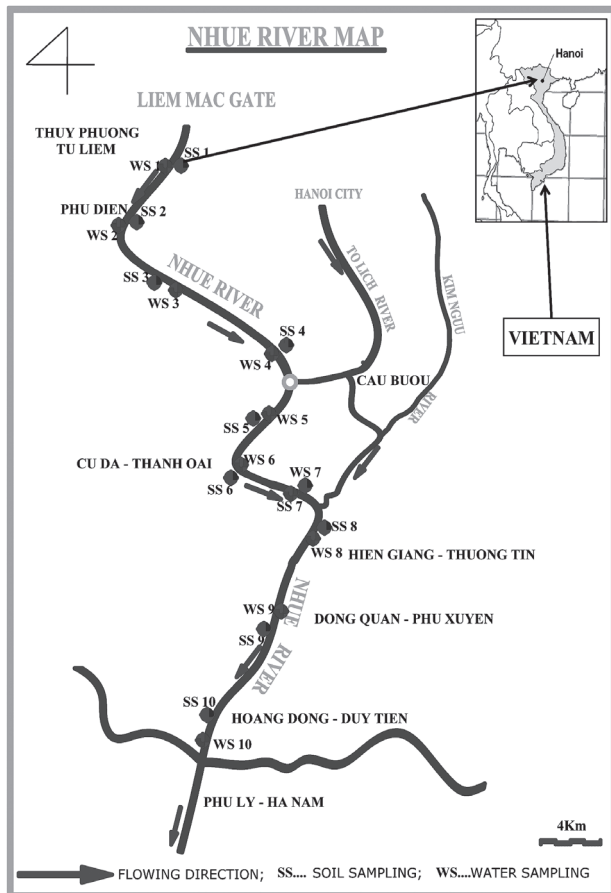


Fig. 1. Site sampling location.

Methodology

Chemical and Physical Properties of Soil

The pH was measured using soil suspension with soil (dry)-to-water ratio of 1:2.5. Organic carbon (OC) contents were determined by Tyurin method, and the organic matter concentration were calculated by multiplying OC values by the coefficient of 1.724 (Committee of Soil Standard Methods for Analyses and Measurements, 1986). Electric conductivity (EC) was determined using conductivity meter (CM 20S TOA). Cation exchange capacity (CEC) and exchangeable cations were measured by the method proposed by Muramoto *et al.* (1992).

In the particle-size analysis, 10g of an air-dried soil/sediment sample was treated with 7% water to remove organic matter, dispersed by ultrasonic vibra-

tion (tank-type; 38 kHz, 250 W), and deflocculated by adjusting pH to 10 with addition of 1 M NaOH. After sedimentation for a fixed period of time, the clay fraction ($< 2\mu\text{m}$) was siphoned out. With repetition of sonication (50T)-sedimentation-siphoning with intermittent pH adjustment, the whole clay fraction was separated. The silt fraction ($2\text{--}20\mu\text{m}$) was also separated by repeated sedimentation and siphoning. The fine sand ($20\text{--}200\mu\text{m}$) and coarse sand ($200\text{--}2000\mu\text{m}$) fractions were obtained by wet sieving. After oven-drying at 105°C , each fraction was weighed to calculate the particle size distribution of a soil/sediment.

For clay mineral analysis, the $< 2\mu\text{m}$ clay fractions of the soil/sediment separated in the particle-size analysis was used. Two clay suspensions containing 50 mg clay were prepared. One was saturated with Mg^{2+} by washing with 0.5 M MgCl_2 and the other with K^+ by 1 M KCl. Excess salt was removed by washing with water. After addition of 1 mL of water was added and an aliquot of suspension containing 30 mg of clay was deposited onto a glass slide, air-dried, and X-rayed. The K-saturated specimen was heated at 300°C and 550°C for 1 hr, and measured after each treatment. The Mg-saturated specimen was solvated with glycerol before a further analysis. The specimen thus treated was X-rayed under 10 mA and 30 kV. Filtered $\text{CoK}\alpha$ or $\text{CuK}\alpha$ radiation from Rigaku diffract meter were used for X-ray diffraction. Relative mineral contents in the clay fractions were estimated based on the peak intensities of XRD (Nguyen and Egashira, 2005).

Total Heavy Metal Concentration of Water

Total heavy metal concentrations were determined for non-filtered water samples, respectively. Heavy metals in the water samples were analyzed with atomic absorption spectrophotometer (AAS – Solar S2 Thermo electronic cooperation).

Total Heavy Metal Concentration of Soil

For total heavy metal concentration, one g of air-dried soil/sediment sample was digested with 5 mL of concentrated HNO_3 . The mixture was brought to a slow boil by heating the flask at 96°C on a hot plate for 1 hr. After the mixture was boiled down to about 2 mL, 5 mL concentrated HNO_3 was added. A reflux cap was placed onto the flask, and the mixture was boiled for approximately 2 h. The solution obtained after centrifugation was transferred into 50 mL volumetric flask and made up to the mark with distilled water after cooling. The solution was analyzed for Cd, Cr, Cu, Ni, Pb and Zn by atomic absorption spectrometry (SOLAAR m, Nippon Jarrel-ash Co., Ltd., Japan). The determination was made in duplicate and the relative deviation of the duplicate values was usually less than 5%.

RESULTS AND DISCUSSIONS

The Quality of Water

Water from the Nhue River is used to irrigate agricultural land in surrounding area of Hanoi city. The irri-

gationwater is heavily polluted with wastes from domestic (60–80%) and industrial (20–40%) source (Klank *et al.*, 2006). Table 1 shows the mean value (four times taking sample) of heavy metal concentration of the water irrigated to the paddy field used for the present study. The total concentrations of heavy metal of almost irrigation water samples (except water samples were taken from upstream and downstream) exceeded the

permissible level of the Vietnamese standard for irrigation water (MOSTE, 2008).

The Quality of the Soil

Chemical and Physical Properties of Soil

Table 2 shows the mean value of chemical and physical properties of soil samples (four times taking sample). The $\text{pH}_{(\text{H}_2\text{O})}$ and $\text{pH}_{(\text{KCl})}$ were in a range of 5.8–6.4 and 5.5 to 6.0, respectively, suggesting the natural condition of soils was lightly acid. The EC ranged between 0.05 and 0.13 mS/cm. The organic matter content varied from 1.4 to 3.6%. Those values were lower than for the agricultural soil along the To Lich and Kim Nguu Rivers studied by Nguyen *et al.* (2010a, 2008b). The soil samples from SS1 exhibited the highest organic matter content. The cation exchange capacity (CEC) ranged between 16.9 and 26.4 cmol_e/kg. The exchangeable cations were predominant in Ca, followed by Mg, K and Na.

The clay, silt, fine sand and coarse sand fractions ranged from 36.5 to 48.3%, 25.1–35.1%, 8.9–15.2% and 11.6–21.8%, being classified as Clay loam (ISSU). These clay contents are higher than those for the soil samples taken from farmland along the To Lich and Kim Nguu Rivers (Nguyen *et al.*, 2010a, 2008b).

The XRD patterns for the < 2 μm clay fractions of the soil samples are illustrated in Fig. 2. The presence of mica was indicated by the 1.00 nm peak along its higher-order reflections at 0.50 and 0.33 nm. The presence of kaolinite was indicated by the peaks at 0.72 and 0.35 nm in the K-saturated specimen that disappeared at 550°C. Chlorite was identified by the peaks at 1.43, 0.72, 0.47 and 0.35 nm. Vermiculite was identified by the peaks at 1.43 nm in Mg-saturated and Mg-glycerol specimens, and the peak at 1.00 nm in the K-saturated spec-

Table 1. Heavy metal concentrations of water of the Nhue River

No	Total heavy metal concentration Dry season (mg/L)					
	Cd	Cr	Cu	Ni	Pb	Zn
WS1	0.009	0.03	0.2	0.07	0.2	1.1
WS2	0.09	0.09	0.4	0.09	0.3	1.3
WS3	0.03	0.07	1.0	0.12	0.6	1.4
WS4	0.06	0.07	1.0	0.17	0.8	1.6
WS5	0.06	0.07	0.9	0.25	1.1	1.7
WS6	0.05	0.08	0.9	0.20	0.9	1.5
WS7	0.04	0.07	0.8	0.15	0.8	1.1
WS8	0.05	0.06	0.9	0.23	0.8	1.5
WS9	0.03	0.05	0.6	0.20	0.6	1.6
WS10	0.01	0.03	0.3	0.15	0.3	1.2
MOSTE**	0.01	0.04	0.5	0.1	0.5	1.5
WHO***	0.01	0.1	0.2	0.2	5	2

* pH was measured in a laboratory

** **MOSTE (08:2008 B1)**: applied to the surface water used for the purpose other than domestic water supply, including irrigation water

*** Irrigation water standard (WHO, 1989)

Table 2. Chemical and physical properties of the soil samples

Location	$\text{pH}_{(\text{H}_2\text{O})}$	$\text{pH}_{(\text{KCl})}$	EC (mS/cm)	OM* (%)	CEC (cmol/kg)	Exchangeable cation (cmol/kg)				Particle size distribution (%)				Texture**
						Ca	Mg	K	Na	Clay (< 2 μm)	Silt (2–20 μm)	Fine sand (20–200 μm)	Coarse sand (200–1,000 μm)	
SS 1	6.2	5.9	0.05	3.6	26.4	17.0	2.2	0.43	0.12	37.3	28.7	13.7	20.3	CL
SS 2	5.8	5.7	0.06	2.8	20.1	15.0	2.0	0.47	0.16	36.5	29.4	15.2	18.9	CL
SS 3	6.1	6.0	0.06	2.4	19.2	13.2	2.5	0.51	0.18	37.2	28.5	14.3	20.0	CL
SS 4	6.1	5.9	0.07	2.0	18.0	12.5	2.4	0.56	0.14	38.5	26.5	13.2	21.8	CL
SS 5	6.4	5.8	0.08	1.6	16.7	11.3	3.3	0.62	0.23	44.1	26.6	12.1	17.2	CL
SS 6	6.2	5.5	0.07	1.7	18.5	15.0	2.9	0.55	0.24	42.1	25.1	13.2	19.6	CL
SS 7	6.6	5.8	0.07	1.8	22.9	17.6	2.6	0.52	0.26	42.8	34.5	10.0	12.7	CL
SS 8	6.2	5.8	0.08	1.4	16.9	14.0	3.7	0.52	0.35	48.3	31.2	8.9	11.6	CL
SS 9	6.2	6.0	0.09	2.0	19.2	13.2	3.2	0.54	0.31	45.6	28.5	9.3	16.6	CL
SS 10	5.8	5.5	0.13	2.6	22.9	17.3	3.6	0.63	0.21	40.8	25.1	15.6	18.5	CL

*OM: Organic Matter Content

**Texture: CL: Clay Loam

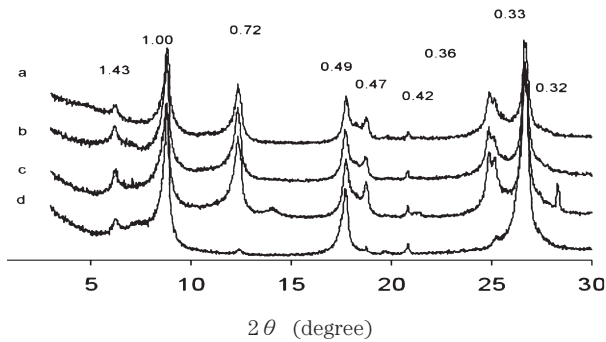


Fig. 2. X-ray patterns of the < 2 μm clay fraction of sediment (SD 4). Spacing is in nm.

Treatments: a, Mg-saturation and glycerol – salivation;
b, Mg-saturation and air-drying;
c, K-saturation and air-drying;
d, K-saturation and heating at 500°C

Table 3. Clay mineral contents in < 2 μm clay fractions of the soil samples

Location	Clay mineral contents (%)				
	Mc*	Ch	Kt	Qz	Fd
SS 1	30	12	23	30	5
SS 2	30	10	21	39	+
SS 3	31	11	20	38	+
SS 4	38	9	19	42	+
SS 5	45	7	18	30	+
SS 6	47	5	14	34	+
SS 7	50	6	17	27	+
SS 8	40	10	20	25	5
SS 9	42	9	20	24	5
SS 10	38	11	17	34	0

*. Mc: Mica; Ch: Chlorite; Kt: Kaolinite; Qz: Quartz; Fd: Feldspar

+. Not detected

imen. The peaks at 0.42 and 0.33 nm and at around 0.32 nm indicate quartz and feldspar, respectively.

The relative percentage of clay minerals of soil samples was calculated based on the peak intensity of the minerals (Nguyen and Egashira, 2005) and the results are given in Table 3. Mica and quartz were predominant with a range of 30–50% and 27–42%, respectively, followed by kaolinite and chlorite. The mineral composition thus obtained was similar to that reported by Ho *et al.*, (1998) for soil in farmland 15 km distant from our study site. Soils in both farmlands are derived from fine particles transported by the Red River. A similar mineral composition was found in farmland soil along the To Lich and Kim Nguu Rivers (Huong *et al.*, 2010a, 2008b).

Heavy Metal Concentration in the Soil

The mean value of four times taking for one location of total heavy metal concentrations in the soil samples are shown in Table 4. The concentration of the

Table 4. Total heavy metal concentration in the soil samples

Location	Cd	Cr	Cu	Pb	Zn	Ni
	(mg/kg)					
SS 1	1.2	42	22	30	127	26
SS 2	2.6	60	42	49	139	39
SS 3	1.4	49	60	58	150	42
SS 4	1.9	50	62	65	157	57
SS 5	2.4	50	61	86	245	54
SS 6	2.4	49	58	75	225	43
vbSS 7	2.3	46	55	72	203	41
SS 8	2.3	43	55	72	215	51
SS 9	1.8	43	49	60	156	43
SS 10	1.3	42	35	55	119	38
BGL*	1.2	42	43	32	115	34
MOSTE**	2		50	70	200	

*. Background levels of heavy metals in agricultural soils of Tu Liem District of Hanoi, Vietnam

**. MOSTE 2008: Vietnamese standard for heavy metal concentration in agricultural soils

heavy metals of the soil was in the range of 22–62 mg/kg for Cu, 30–86 mg/kg for Pb, 119–245 mg/kg for Zn, 1.2–2.6 mg/kg for Cd, 42–60 mg/kg for Cr, and 26–57 mg/kg for Ni. The metal concentrations tended to increase toward the sampling locations from upstream to the centre of river and then decrease toward to downstream. This would be the result of settlement of heavy metal-sorbed suspended solids in irrigation water during its flowing in channels from the pumping stations to the remote sampling locations.

For the sewage irrigated soil, greater amounts of heavy metals were observed, especially for Cd, Zn, Pb, Cr (Chen *et al.*, 2007). According to Yamadera and Suzuki (2004), soil of Hanoi city general developed on alluvial deposit from Red river. Mica as well as a kaolinite and chlorite dominate clay minerals over the whole region. Thus, there is no big difference in the parent material of soils. Therefore, the great elevation of heavy metal concentrations in soil of study area can be considered to result from using polluted irrigation water.

Compared with the background level of heavy metals in the soil that has not been subjected to polluted irrigation water (Ho *et al.*, 1998), the heavy metal concentration exhibited higher values in all locations of the study site except SS1 for Cu, Pb and Ni, and the metal concentrations for Cd, Cu and Pb of soil samples SS 3 – SS 8 exceeded the permissible level of the Vietnamese standard (MOSTE, 2008) for agricultural soil (Cd: 2 mg/kg; Cu: 50 mg/kg; Pb: 70 mg/kg). These indicate that the excess heavy metal concentrations in the soil are mainly due to the application of polluted irrigation water to the farmland. Yet, metal concentrations in the soil samples were much lower than the concentrations in the farmland irrigated from the heavily polluted To Lich and Kim Nguu Rivers (Nguyen *et al.*, 2008b). In the long term, irrigation would cause the accumulation of heavy

Table 5. Correlation coefficients among the chemical properties of the soil samples

	pH	Clay	CEC	OM	Cd	Cr	Cu	Ni	Pb	Zn
pH	111	0.47	-0.33	-0.48	0.35	0.40	0.37	0.42	0.4	0.36
Clay		1	0.93	0.91	0.95	0.97	0.92	0.96	0.94	0.93
CEC			1	0.86	-0.96	-0.97	-0.97	-0.98	-0.98	-0.99
OM				1	-0.95	-0.86	-0.97	-0.83	-0.91	-0.87
Cd					1	0.96	0.95	0.96	0.98	0.94
Cr						1	0.97	0.99	0.97	0.98
Cu							1	0.98	0.99	0.98
Ni								1	0.98	0.98
Pb									1	0.99
Zn										1

*Correlation coefficient greater than 0.83 is significant at 1% level

metals in the soil at a toxic concentration level and adversely affect both soil microbiological process and plant growth.

Compared with the background level of heavy metals in the soil that has not been subjected to polluted water irrigation (Ho *et al.*, 1998), heavy metals in almost locations of the study site exhibited higher concentration, and the total concentrations of Cd, Cu and Pb exceeded the permissible level of the Vietnamese standard for agricultural soil (MOSTE, 2008). These indicate that the excess heavy metal concentrations in the study site can be attributed mainly to the application of polluted irrigation water to the soil.

From Tables 1 and 4 shows that the higher metal concentrations in the water lead to the higher metal concentration in the soil samples. These results indicate that heavier water pollution leads to higher heavy metal concentration in the soil, and the effect of heavy metal pollution of the water on the agricultural soil is different according the type of heavy metal.

The source of heavily polluted is the water from the To Lich and Kim Nguu Rivers with extremely high suspended solid concentration (Nguyen *et al.*, 2008a). Through irrigation, suspended solids of the Nhue River are transported into the agricultural field, and the related metals accumulated in the soil.

Table 5 shows the correlation coefficients among the chemical properties of the soil samples. The correlation coefficient was significant at 1% level among the chemical properties excepting pH.

CONCLUSIONS

The concentrations of Cr, Cd, Cu, Pb, Zn and Ni in the soil at the sampling sites affected by the polluted water from the Nhue River exceeded the permissible level of Vietnamese standard for agricultural soil. Metals concentration in the soils increased with increasing metal concentrations in water. These results indicate that the heavier pollution of the water led to the higher metal concentration in the soil.

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