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Effects of using wastewater as nutrient sources on soil chemical properties in peri-urban agricultural systems

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Abstract. Reusing domestic wastewater for irrigation and applying biosolids as a fertiliser in crop production are common practices in peri-urban areas of Vietnam. This study investigates the effects of using domestic wastewater in field experiments on Fluvisols soils in peri-urban areas of Hanoi and Nam Dinh cities. We compared long-term (30-50 years) wastewater-irrigated rice-dominated farming systems. Using wastewater for irrigation significantly affected pH, electrical conductivity (EC), exchangeable K and Na and reverse *aqua regia*-digestible (Rev *Aq Reg*) copper (Cu), lead (Pb) and zinc (Zn) in the investigated areas compared with control plots irrigated using river water. There were no significant effects of wastewater irrigation on the NH_4NO_3-extractable fraction of cadmium (Cd) and other trace metals, but the EDTA-extractable fraction of Cu, Pb and Zn was significantly increased.

Keywords: SE Asia; heavy metals; irrigation; paddy soils; trace elements

1. Introduction

Urbanisation and industrialisation are leading to production of a huge volume of effluents in many countries. Industrial, agricultural and domestic effluents such as biosolids and wastewater are either dumped on land or used for irrigation and fertilisation purposes, which creates both opportunities and problems [24].

The advantages of reusing wastewater are that it provides a convenient disposal of waste products and has the beneficial aspects of adding valuable plant nutrients and organic matter to soil. Furthermore, the reuse of wastewater for irrigation as a fertiliser source is a common and popular practice, especially in peri-urban areas. Wastewater is often the only source of water for irrigation. Even in areas where wastewater is not the sole water source for agricultural irrigation, farmers still prefer using sewage for irrigation by reason of its nutritive value, which reduces expenditure on chemical fertilisers [10, 17].

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However, as wastes are products of human society, enhanced concentrations of potential toxic substances including trace metals are generally found in wastewater, which may limit the long-term use of effluents for agricultural purposes due to the likelihood of phytotoxicity, health and environmental effects [1, 14]. Another problem of wastewater disposal on agricultural land is the potentially phytotoxic nature of organic wastes, mainly as a result of combination of factors such as high salinity or excess of ammonium ions, organic compounds or low molecular weight fatty acids, which e.g. may inhibit seed germination [6, 10].

If the content of trace metals increases above a certain critical concentration due to their accumulation in soil, this can have negative environmental effects, which can include negative effects on soil biota and hence on microbial and faunal activity [7]. Furthermore, trace metals can affect crop growth and quality, and thus pose risks for human health [2, 6, 12]. Therefore, the risk of contamination by trace metals must be considered when wastewater is applied and understanding of the behaviour of metals in the soil is essential for assessing environmental risks of applying wastewater in agro-ecosystems.

The main objective of this paper was to quantify the effects of reuse of wastewater as nutrient sources by: (i) investigating the effects of long-term wastewater irrigation on soil pH, EC, organic carbon, total nitrogen and trace metals (cadmium (Cd), copper (Cu), lead (Pb) and zinc (Zn)); (ii) investigating the effects of application of wastewater, especially as regards trace metal accumulation and solubility.

2. Materials and methods

2.1. *Location of the research areas*

Soil samples were collected from peri-urban areas in two provinces of Vietnam, including Hanoi, Nam Dinh (Table 1). The sampled areas are located in delta and lowland areas with a tropical monsoon climate. The annual rainfall is 1500-2000 mm, and more than 50% of the rainfall is concentrated during June to August. The mean monthly temperature varies between 17 and 29°C, with the warmest period from June to August and the coldest during December and January.

Table 1. Description of wastewater (full-scale case studies) in experiments on Fluvisols in peri-urban areas of Hanoi and Nam Dinh cities, Vietnam

No.	Location	Soil irrigated by	Position	Name	Crop	Application	Sampling time
1	Hanoi						
	Fluvisols	Wastewater	N: 20°57.52' E: 105°49.68'	Treatment	Rice	Since 1960s	June 2004
		River water	N: 20°58.12' E: 105°48.15'	Control	Rice		June 2004
2	Nam Dinh						
	Fluvisols	Wastewater	N: 20°44.93' E: 106°20.98'	Treatment	Rice	Since 1980s	June 2004
		River water	N: 20°43.43' E: 106°20.68'	Control	Rice		June 2004

2.2. Wastewater irrigation in peri-urban agricultural production systems

The sewage irrigation study areas are located in urban regions downstream from Hanoi City and Nam Dinh City. The soil types are Eutric Fluvisol at the Hanoi site, and Humi-Endogleyic Fluvisol (Eutric) at the Nam Dinh site according to the World Reference Base for Soil Resources. The soils are fertile and suitable for growing crops. Rice has been the main crop in these areas, but there is a tendency of changing from rice to vegetable production due to increasing demand from the inner city markets of Hanoi and Nam Dinh. At the Hanoi site, sewage water has been used for irrigation since the 1960s. Because of water scarcity, agricultural land has been irrigated by sewage from Kim Nguu River, which runs through the urban area to rural agricultural land [8, 9]. At the Nam Dinh site, irrigation using wastewater started in the 1980s as a result of increasing urbanisation. The sewage mainly comprises domestic water but also includes wastewater and discharges from industrial activities in the urban areas [4, 20]. In Nam Dinh, the soil samples were taken in the fields where the DANIDA-IWMI project on wastewater reuse in agriculture in Vietnam was carried out [20].

2.3. Soil sampling strategy and sample preparation

For assessment of the impact of wastewater, soil samples were taken from the topsoil (0-20 cm) of all study sites in peri-urban areas of Hanoi (n=4) and Nam Dinh (n=8) using a soil auger. At every sampling point, 3 to 5 sub-samples were taken from approximately 250 m^2 and mixed to obtain a bulk sample. Non-wastewater irrigated soils ("natural" river irrigation) were also sampled for comparison (n=4 for Hanoi, and n=8 for Nam Dinh).

After air drying at room temperature, the soil samples were ground and sieved to remove particles > 2 mm, and then stored in plastic bags. The soil samples were brought to Sweden (SLU) for analysis.

2.4. Soil analysis

Total N (N_{tot}) and total organic carbon (TOC) was determined on finely ground samples on a LECO CHN analyser (Leco CHN®CHN 932 analyser). Prior to the analyses, the samples were treated by 4M HCl (1:1 soil:solution ratio) for dissolution of carbonates. The soil EC and pH were measured in deionised H_2O (1:5 soil:solution ratio), and pH_{CaCl_2} was determined after addition of 0.5M $CaCl_2$ [18]. The soil samples were extracted with 1M NH_4NO_3 for 2 hours (1:2.5 soil:solution ratio) to quantify the exchangeable and specifically adsorbed fraction of trace metals (*i.e.* Cd, Cu, Pb, Zn) [3]. Potentially bioavailable metals were extracted with 0.025 M $(Na)_2$EDTA (1:10 soil:solution ratio) for 1.5 h [19]. The reverse *aqua regia* (3:1 HNO_3:HCl ratio)-digestible fraction (Rev *Aq Reg*) of Cd, Cu, Pb and Zn was extracted using a method described by Stevens *et al.* [18]. After centrifugation, filtration and dilution (if necessary) metal concentrations were determined by inductively coupled plasma-mass spectrometry (ICP-MS, Perkin Elmer ELAN 6100).

2.5. Water sampling and water analysis

In Nam Dinh and Hanoi, water samples were collected in summer 2004 from the Red River and wastewater channels, which were the irrigation sources at the study sites. The pH and EC in these water samples were determined directly after sampling. Polyethylene bottles that had been prewashed with acid and distilled water and dried were used, and after sampling, a few drops of concentrated HCl were added prior to chemical analysis.

Water samples were analysed for their total concentrations of Ca, Cd, Cu, K, Mg, N, P, Pb and Zn. One aliquot of the samples was digested with boiling concentrated HNO_3 before determination of the total concentration of K by flame spectrometry; Ca, Mg and Na, by atomic adsorption spectrophotometry (AAS, Perkin Elmer 300); Cd, Cu, Pb and Zn by ICP-MS, and total P by HNO_3 digestion followed by determination of PO_4-P with the ascorbic acid method [5]. Total N was quantified as described elsewhere [9].

2.6. *Statistical analysis*

Data from the experiments were analysed using the General Linear Model (GLM) procedure of Minitab Software version 14. Treatment means which showed significant differences at the probability level of $P<0.05$ were compared using Tukey's pairwise comparison procedure. The source of irrigation water within sites (wastewater and river water) was used as a factor in the model. The statistical model used was $y_{ij} = \mu + \alpha_i + e_{ij}$, where μ is the mean value for all treatment, α_i the different between mean value of treatment i with overall mean, and e_{ij} is the random error.

3. Results

3.1. *Irrigation water quality*

The results of the irrigation water analysis are presented in Table 2. The pH and EC were significantly higher in the wastewater compared with the river water. The wastewater also had significantly higher concentrations of nutrients and trace metals compared with the river water. This indicated that non-treated wastewater contained both nutrients that are of value for irrigation of crops in agricultural systems, and potential toxic elements that can affect soil production capacity and crop quality. A comparison between wastewaters in Hanoi and Nam Dinh showed that the concentrations in Hanoi wastewater were significantly higher for most elements included in the study (*i.e.* Cd, Cu, K, Na, N_{tot}, P_{tot}, Pb, Zn).

3.2. *Effects of applying wastewater on soil pH and electrical conductivity*

Applying wastewater for irrigation significantly increased soil pH (pH_{H2O} and pH_{CaCl2}) at both study sites (Hanoi and Nam Dinh) (Table 3), probably due to wastewater being more alkaline than river water (Table 2). The similar effect was observed for electrical conductivity, which was higher in the wastewater treatments than in the control (river).

3.3. *Effects of applying wastewater on soil organic carbon and total nitrogen contents*

Reuse of wastewater for irrigation caused an increase in total organic carbon (TOC) content and total nitrogen (N_{tot}) in the soil at both study sites (Fig. 1). The soils that had received wastewater for irrigation had 1.68% TOC and 0.19% N_{tot} at the Hanoi site and 2.67% TOC and 0.26% N_{tot} at the Nam Dinh site. The corresponding values for control samples were 1.29% (TOC), 0.15% (N_{tot}) and 1.85%, (TOC), 0.21% (N_{tot}) for Hanoi and Nam Dinh, respectively.

Table 2. Water quality of the Red River water and wastewater used for irrigation in Hanoi and Nam Dinh. Different letters (a, b) denote significant differences between sources of irrigation water within sites ($P<0.05$)

No	Parameter	Units	Nam Dinh		Hanoi	
			Red River (n=4)	Wastewater (n=5)	Red River (n=4)	Wastewater (n=6)
1	pH		7.1^{a}	8.0^{b}	6.9^{a}	7.9^{b}
2	EC	dS m^{-1}	0.20^{a}	0.82^{b}	0.21^{a}	0.86^{b}
3	Total nitrogen (N_{tot})	mg L^{-1}	4.1^{a}	10.8^{b}	3.9^{a}	19.2^{b}
4	Total phosphorus (P_{tot})	mg L^{-1}	0.6^{a}	2.0^{b}	0.5^{a}	4.4^{b}
5	Potassium (K)	mg L^{-1}	3.1^{a}	6.8^{b}	4.1^{a}	12.8^{b}
6	Sodium (Na)	mg L^{-1}	32.5^{a}	85.5^{b}	28.6^{a}	135.7^{b}
7	Calcium (Ca)	mg L^{-1}	46.7	48.9	54.6	54.3
8	Magnesium (Mg)	mg L^{-1}	12.5	10.1	14.2	12.4
9	Lead (Pb)	μg L^{-1}	1^{a}	2^{b}	2^{a}	3^{b}
10	Zinc (Zn)	μg L^{-1}	32^{a}	67^{b}	24^{a}	236^{b}
11	Copper (Cu)	μg L^{-1}	14^{a}	42^{b}	18^{a}	82^{b}
12	Cadmium (Cd)	μg L^{-1}	0.2^{a}	0.5^{b}	0.5^{a}	0.9^{b}

Table 3. Electric conductivity (EC, μS cm^{-1}), pH, exchangeable Ca, Mg, Na, K (1M NH_4NO_3 extractable; g kg^{-1}) in topsoil (0-20 cm) samples from experiments with reuse of wastewater. Different letters denote significant differences between treatments at the same site ($P<0.05$)

Site	EC[1]	pH^{1}_{H2O}	pH^{2}_{CaCl2}	Exchangeable[3]			
				K	Na	Ca	Mg
Hanoi							
Control	62.80^{a}	6.45^{a}	5.69^{a}	0.06^{a}	0.04^{a}	0.89	0.19
Wastewater	102.75^{b}	6.70^{b}	5.96^{b}	0.16^{b}	0.09^{b}	1.00	0.22
Nam Dinh							
Control	78.25^{a}	5.99^{a}	5.42^{a}	0.06^{a}	0.04^{a}	1.55	0.18
Wastewater	179.38^{b}	6.36^{b}	5.71^{b}	0.12^{b}	0.17^{b}	1.60	0.21

[1] *pH in H_2O, ratio soil: water = 1: 5*
[2] *pH in 0.05 M $CaCl_2$, ratio soil: solution = 1:5*
[3] 1M NH_4NO_3 extractable, ratio soil: solution = 1:2.5

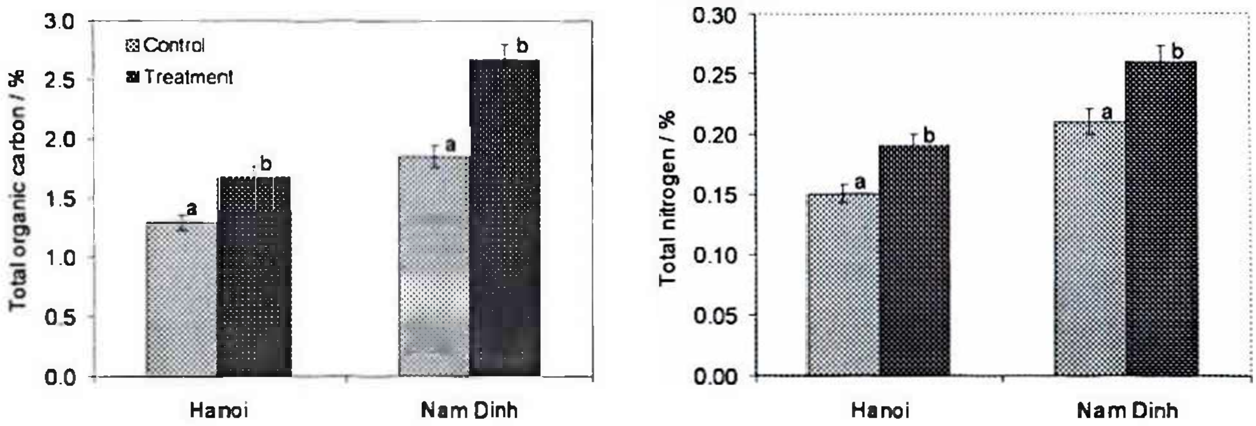

Fig. 1. Effect of wastewater irrigation on soil chemical properties, total organic carbon (TOC, %), total nitrogen (N_{tot}, %). Different letters denote significant differences between treatment and control at the same site ($P<0.05$).

3.4. Effects of applying wastewater on trace metal concentrations in soil

The concentrations of reverse *aqua regia* (Rev *Aq Reg*)-digestible Cu, Pb and Zn in soils receiving wastewater were significantly higher than those in soils receiving river water. There was no significant difference in Cd concentration (Rev *Aq Reg*) between wastewater irrigated soils and control soils (Fig. 3). The potentially bioavailable concentrations of Cu, Pb and Zn (EDTA-extractable) in wastewater-treated soils were significantly higher than in control soils (no difference for Cd). The NH_4NO_3-extracted fractions of Cd, Cu, Pb and Zn constituted only a small proportion of the EDTA-extracted fractions. However, there was no significant difference between treated soils and control soils in the exchangeable (NH_4NO_3) fraction of these metals (Table 4). The reason of this might be low concentrations in combination with a variation between the replicates.

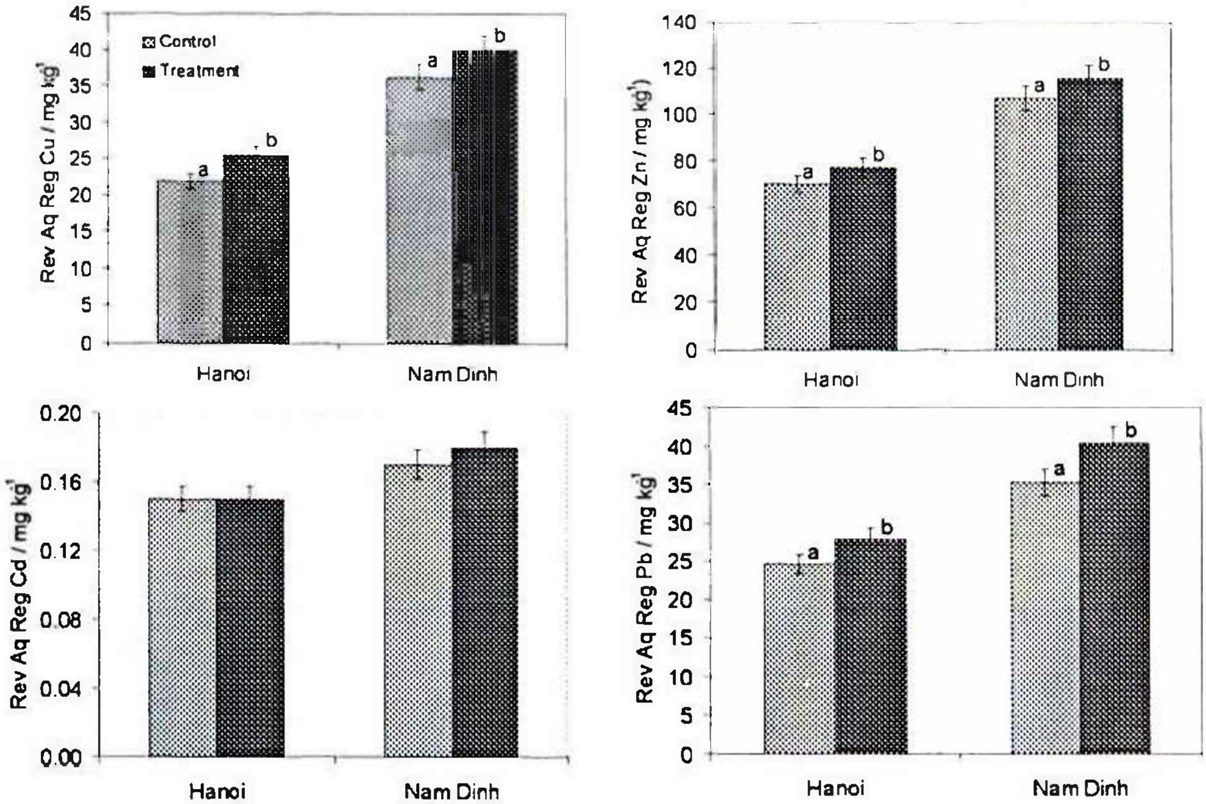

Fig. 2. Effect of reuse of wastewater on reverse *aqua regia* (Rev *Aq Reg*)-extractable Cd, Cu, Pb and Zn (mg kg^{-1}) concentrations in soil. Different letters denote significant differences between treatment and control at the same site ($P<0.05$).

Table 4. Effect of wastewater application on 0.025 M EDTA (mg kg^{-1} dw) and 1M NH_4NO_3 extractable (mg kg^{-1} dw) trace metals. Different letters denote significant differences between treatment and control at the same site ($P<0.05$)

Site	EDTA-extractable				NH_4NO_3-extractable			
	Cu	Zn	Cd	Pb	Cu	Zn	Cd	Pb
Hanoi								
Control	8.21^a	1.85^a	0.112^a	7.18^a	0.002	0.150	0.0059	0.004
Treatment	9.53^b	2.97^b	0.105^a	8.89^b	0.004	0.205	0.0076	0.007
Nam Dinh								
Control	10.99^a	1.63^a	0.120^b	11.10^a	0.009	0.120	0.0093	0.011
Treatment	12.65^b	1.75^b	0.126^b	15.32^b	0.006	0.180	0.0125	0.038

4. Discussion

Analyses of soil samples collected to assess the impacts of sewage irrigation on the irrigated agricultural soils of peri-urban areas of Hanoi and Nam Dinh cities showed that reuse of municipal wastewater for irrigation had significantly increased both TOC and N_{tot} in soils. This finding is in agreement with previous studies where wastewater irrigation had been shown to increase soil organic C and N [15]. However, a potential hazard to peri-urban crop production was revealed due to the accumulation of trace metals in agricultural soils irrigated with sewage.

Municipal wastewater contains a variety of inorganic substances from domestic and industrial sources, including a number of potentially toxic elements such as arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), lead (Pb), zinc (Zn), *etc.* [16]. According to the annual report on the environmental status of Vietnam made by VEPA [22], these potential toxic elements are commonly present in domestic wastewater of many cities in Vietnam. Even if potential toxic elements in wastewater are not present in concentrations likely to directly affect humans and thus limit their agricultural use, they seem to be higher than in natural river water, which would contaminate the agricultural soils in the long-term. As a result, the concentrations of trace metals (Cu, Pb and Zn) in the wastewater-irrigated soils were significantly higher than in control soils, indicating that the application of wastewater had enriched the soil with trace metals. Liu *et al.* (2005) studied the impact of sewage irrigation on trace metal contamination in Beijing and reported that the trace metals were enriched in the soil due to sewage irrigation [11]. This was also found in earlier publications about effects of sewage irrigation on soils [13, 21].

The application of wastewater in the peri-urban sites of Hanoi and Nam Dinh cities increased soil pH by approximately 0.3 units compared with the non-wastewater irrigated sites. Previous researches [8, 23] have indicated that the wastewater applied for irrigation at Hanoi and Nam Dinh sites is in most cases neutral to alkaline (6.5-8.5). The present study also found that the pH was significantly higher for wastewater compared with natural river water (Table 2). In addition, the higher concentration of cations such as Na and K in wastewater led to an increase in EC and exchangeable Na and K in soils irrigated with wastewater. The high pH of soils irrigated with wastewater might reduce the mobility of the trace metals accumulated in these soils.

5. Conclusions

Reuse of wastewater as nutrient sources has become common practice in Vietnam, especially in peri-urban areas. The reuse of these nutrients had some beneficial effects on soil fertility, such as increased total organic carbon and nitrogen. This study found that both organic carbon content and total nitrogen were improved (increased) in soils treated with wastewater. However, these benefits were limited by the presence of some potential toxic trace metals in wastewater. It was concluded that the reuse of wastewater for irrigation increased soil pH, EC, TOC, N_{tot} and total concentration of Cu, Zn and Pb. The EDTA-extractable fraction of Cd, Cu and Zn was significantly higher for wastewater-irrigated soils.

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