Effects of Different Patterns of Land Use on Status of Heavy Metals in Agricultural Soils

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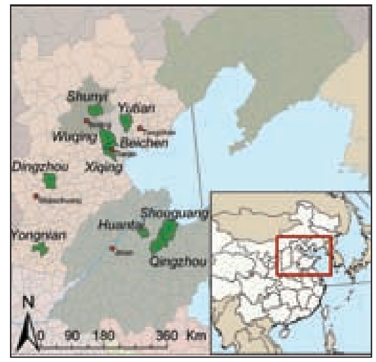
Long-term use of high rates of chemical fertilizers and organic manures in open vegetable fields and field-scale greenhouse vegetable production contributed to the accumulation of Cu and Zn, while changes for other heavy metals were not detected. The contents of total Cu, Zn, and other heavy metals in soils increased with vegetable production history.



Farmers in China use much higher nutrient application rates in their vegetable production systems compared to grain production systems. Long-term and excessive fertilization, especially in the case of P fertilization and organic manure use, can increase the risk for serious heavy metal pollution. Vegetable production within the environment of suburban China can also be easily affected by the waste air, water, and residue from industry, plus exhaust emitted by automobiles. Water and soils in the suburbs have suffered from heavy metal pollution to some extent, which might lead to excessive accumulation of one or several heavy metals, such as Cd, Hg, Cr, As, and Pb in vegetables (Huang et al., 2007).

Less information is available on the effect of long-term use of high rates of chemical fertilizers and organic manures on accumulation of heavy metals in more rural regions, especially in vegetable production areas. The objective of this study was to analyze heavy metal contents and their contamination status in agricultural soils under three typical land use practices to provide scientific basis for improving environmental quality in these agricultural soils and fertilization techniques for high yield and high quality crop production.

Table 1. Environmental Quality Standard for Soils (GB15618-1995; mg/kg, weight of air-dried soil).

Item	Background values of uncultivated soil	The 2nd criterion of Environmental Quality Standard for Soils		
		pH < 6.5	pH 6.5-7.5	pH > 7.5
Cu	35	50	100	100
Zn	100	200	250	300
Cd	0.2	0.3	0.3	0.6
Pb	35	250	300	350
Cr	90	150	200	250
As	15	40	30	25
Hg	0.15	0.3	0.5	1

Source: Xia, 1996.

Rural locations that were a significant distance from suburban areas were selected from 10 counties (or districts or cities) in four provinces or municipalities within the Huabei plain in northern China. The selected land use patterns included open vegetable fields (fields under open air), greenhouse vegetable fields (under large-scale plastic greenhouses), and grain crop fields. Specifically, the experimental regions were Shunyi district of Beijing municipality; Xiqing, Beichen and Wuqing districts of Tianjin municipality; Dingzhou city, Yongnian and

The three land use patterns selected for this study included plastic-covered greenhouse vegetables (lower left and right), open vegetable fields (upper left), and grain crop fields (upper right).

Abbreviations and notes: Cu = copper; Zn = zinc; Cd = cadmium; Pb = lead; Cr = chromium; As = arsenic; Hg = mercury; P = phosphorus.

Table 2. Contents of heavy metals in rural soils under different patterns of land use.

Heavy metal	Item	Open vegetable fields	Greenhouse vegetable fields	Grain crop fields
Cu	Range, mg/kg	9.4-73	12.9-81	6.7-41
	Mean, mg/kg	27.7 a	29.9 a	22.3 b
	C.V., %	46	49	38
	Samples > background levels, %[1]	24	25	11
Zn	Range, mg/kg	28-189	38-223	21-112
	Mean, mg/kg	72 a	82 a	57 b
	C.V., %	54	50	40
	Samples > background levels, %	21	20	7
Cd	Range, mg/kg	0.3-1.1	0.3-1.0	0.2-0.9
	Mean, mg/kg	0.6 a	0.6 a	0.6 a
	C.V., %	29	27	27
	Samples > background levels, %	100	100	100
Pb	Range, mg/kg	13-55	15-57	11-49
	Mean, mg/kg	30 a	29.4 a	28 a
	C.V., %	34	35	32
	Samples > background levels, %	26	20	22
Cr	Range, mg/kg	40-128	44-154	35-121
	Mean, mg/kg	83 a	88 a	79 a
	C.V., %	22	24	28
	Samples > background levels, %	26	52	31
As	Range, mg/kg	3-13	3-10	2-11
	Mean, mg/kg	7 a	7 a	6 a
	C.V., %	30	24	31
	Samples > background levels, %	0	0	0
Hg	Range, mg/kg	0.1-0.3	0.1-0.2	0.1-0.2
	Mean, mg/kg	0.1 a	0.1 a	0.1 a
	C.V., %	40	34	39
	Samples > background levels, %	40	28	29

Means with the same letter are not significantly different at $p < 0.05$.

Yutian counties of Hebei province; Shouguang and Qingzhou cities, and Huantai county of Shandong province.

Three to six sampling areas for each land use practice were selected from each of the 10 investigated regions. Each sampling area was about 20 ha. Composite soil samples from a total of 38 open vegetable fields, 40 greenhouse vegetable fields, and 45 grain crop fields were collected using a stainless steel drill from the soil surface layer (0 to 20 cm) between June 18 and 26, 2007. All soil samples were air-dried and ground through a sieve (2 mm for soil pH and 0.149 mm for heavy metals) prior to analysis. The soils were classified as Eutric Cambisols and Haplic Luvisols (FAO, 1988). Soil pH was 7.6±0.8, 7.2±1.0, and 7.8±0.7 for the open vegetable fields, greenhouse vegetable fields, and grain crop fields, respectively. Production history ranged from 5 to 20 years for the open vegetable fields and greenhouse vegetable fields which were previously cropped to wheat and corn, and the grain crop fields had production histories of over 20 years. The main vegetable crops were cabbage, Chinese cabbage, Welsh onion, and eggplant for the open vegetable fields, while tomato, cucumber, Chinese celery, and watermelon were grown on the greenhouse vegetable fields. The most common grain crops were winter wheat and summer corn.

Information of crop production history, including varieties, rotations, and chemical fertilizer and organic manure use from 2005 to 2007 was collected for all of the soil sampling sites mentioned above. Nutrient application rates for one crop season were averaged from 2005 to 2007. Annual nutrient application rates were considered to be the total amount of the nutrient applied for the first and second crop each year (first and second vegetables in open and greenhouse vegetable fields, and the wheat-corn rotation).

The soil samples were digested using aqua regia (HCl/HNO_3, 3:1 solution)-$HClO_4$ (Lu, 2000), and the concentrations of total Cu, Zn, Cd, Pb, and Cr were measured by atomic absorption spectroscopy. The soil samples were digested by a 1:1 HCl/HNO_3 solution for total As (Lu, 2000) and a 2:1 HCl/HNO_3 solution for total Hg (Fan, 2003), and the concentrations of the two elements were determined by atomic fluorescence spectroscopy. Soil pH was measured in a 2.5:1 water:soil suspension using a glass pH electrode. **Table 1** provides the background values for heavy metals in uncultivated soil and the Environmental Quality Standard for Soils (GB15618-1995) taken as assessment criteria for soil heavy metal status (Xia, 1996).

Significantly higher contents of total soil Cu and Zn were found in open vegetable fields and greenhouse vegetable fields compared to grain crop fields (**Table 2**). However, contents of total soil Cd, Pb, Cr, As, and Hg for open vegetable fields and greenhouse vegetable fields did not differ statistically from levels measured within grain crop fields. Obvious differences were observed for the percentage of soil samples having heavy metals contents beyond the assessment criteria. All samples had Cd contents beyond background values for uncultivated soil, while 60 to

Table 3. Fertilizer application rates (kg/ha/year) averaged from 2005 to 2007 for rural soils under different patterns of land use.

Nutrient	Item	Open vegetable fields	Greenhouse vegetable fields	Grain crop fields
N	Fertilizer	650±350	800±520	440±190
	Manure	230±330	670±580	20±70
	Total	880±370	1,470±830	460±170
P_2O_5	Fertilizer	330±290	730±660	170±90
	Manure	210±290	620±540	20±70
	Total	540±340	1,350±880	190±90
K_2O	Fertilizer	260±200	650±720	90±80
	Manure	160±210	390±330	10±50
	Total	420±250	1,040±880	100±90

Table 4. Contents of heavy metals in rural soils under different vegetable production history.

Heavy metal	Land use pattern	5 to 10 years: Number of sampling sites	5 to 10 years: Mean, mg/kg	11 to 20 years: Number of sampling sites	11 to 20 years: Mean, mg/kg
Cu	Open[1]	17	22.7±9.1	21	31.7±14.3
	Greenhouse[2]	22	27.6±10.4	18	32.7±18.6
Zn	Open[1]	17	56.5±19.5	21	84.7±45.8
	Greenhouse[2]	22	76.9±38.3	18	89.1±44.6
Cd	Open[1]	17	0.61±0.19	21	0.64±0.18
	Greenhouse[2]	22	0.63±0.19	18	0.65±0.16
Pb	Open[1]	17	29.0±9.1	21	31.6±11.5
	Greenhouse[2]	22	29.2±10.4	18	29.5±10.5
Cr	Open[1]	17	80.6±14.6	21	84.3±21.1
	Greenhouse[2]	22	82.9±19.0	18	94.0±22.8
As	Open[1]	17	6.4±1.4	21	7.2±2.4
	Greenhouse[2]	22	6.6±1.6	18	6.6±1.6
Hg	Open[1]	17	0.12±0.05	21	0.14±0.05
	Greenhouse[2]	22	0.12±0.04	18	0.15±0.05

[1]Open vegetable field; [2]Greenhouse vegetable field.

80% of samples were above the Environmental Quality Standard for Cd. The percentages of soil samples with Cu, Zn, Pb, Cr, As, and Hg contents beyond reported background values were lower, with values ranging between 0 to 53%, and the contents of all six heavy metals were below the Environmental Quality Standard.

Previous reports list the main sources of heavy metal pollution in agricultural soils as effluent of waste air, water, and residue from industry, auto exhaust, sewage irrigation, and the use of agrochemical materials (Zhu and Zhou, 1999; Zheng et al., 2006). In this study, the selected farmlands (which were considerably more rural), were observed to not be affected by the list of pollution sources above, and no sewage irrigation was found within these experimental regions. Since heavy metals can naturally occur within some P fertilizers, and relatively high contents of Cu, Zn, and other heavy metals occur in organic manures (Nicholson et al., 2003), continuous and combined use of these nutrient sources at high application rates can lead to the accumulation of heavy metals in agricultural soils. Application rates of N, P_2O_5, and K_2O from fertilizer and manures varied considerably with each cropping system, but were noted to be much higher in the selected open vegetable and greenhouse vegetable fields compared with grain crop fields (**Table 3**). Average respective total application rates for the bare vegetable, greenhouse vegetable, and grain crop fields were 882, 1,474, and 455 kg/ha/year for N with 26%, 46%, and 4% of each total originating from manure; 538, 1,349, and 188 kg/ha/year for P_2O_5 with 38%, 46%, and 8% coming from manure; and 416, 1,036 and 103 kg/ha/year for K_2O with 38%, 37%, and 12% coming from manure, respectively. The significantly higher accumulation of soil Cu and Zn for open vegetable and greenhouse vegetable fields in this study are likely a result of both higher application rates from fertilizers and manures, as well as higher proportions of manure application in these systems. However, vegetable production history is also a significant factor. Contents of total soil Cu, Zn, Cd, Pb, Cr, As, and Hg (especially Cu and Zn), increased with vegetable production history (**Table 4**). Under production histories between 5 to 10 years and 11 to 20 years, the respective total soil Cu contents were 22.7 and 31.7 mg/kg for open vegetable fields, and 27.6 and 32.7 mg/kg for greenhouse vegetable fields. Similarly, total soil Zn contents for the two production history ranges were 56.5 and 84.7 mg/kg for open vegetable fields, and 76.9 and 89.1 mg/kg for greenhouse vegetable fields.

These data indicate that soil Cd in all three land use patterns is a potential threat to the food production chain. Although the contents for the remaining six heavy metals were below recognized soil assessment criteria, trends suggest that current vegetable management practices are significantly affecting soil Zn and Cu under both vegetable production systems. BC

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Acknowledgments

This research was supported by the earmarked fund for Modern Agro-industry Technology Research System and the IPNI China Program (Project #CAAS-NMS02).

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