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Potential risk of cadmium in a soil-plant system as a result of long-term (10 years) pig manure application

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ABSTRACT

Animal manure may be a primary source of cadmium (Cd) to Chinese farmlands because abnormally high values of Cd were observed in various manures. In this study, we evaluated the potential risk of Cd in soil-plant (maize and soybean) system as a result of the long-term (10 years) application of pig manure (PM). During 10 years, the loading rate of Cd through PM application ranged from 26.33–131.50 g/ha/year, while the crops removal rate of Cd was relatively small in comparison to the quantity of Cd supplied by PM application (1.03–4.36 g/ha/year). The PM application significantly increased total Cd concentration in soil. Although the Cd levels did not exceed the Chinese soil quality criteria (1.0 mg/kg dry matter (DM)), it would only take less than two years to reach this limit at high PM application rate. The same trend was also observed for the Cd concentration in maize and soybean. More seriously, Cd concentration in grain of soybean was higher than the threshold values for animal and human ingestion (0.2 mg/kg DM). Based on a mass balance calculation, we found that atmospheric deposition was also an important source of Cd in the experimental area (10.27 g/ha/year). Moreover, the application of PM enhanced the leaching loss of Cd, but they were still fairly small (0.34–0.73 g/ha/year).

Keywords: long-term field experiment; contamination; *Glycine max* L.; *Zea mays* L.; fertilization

Cadmium (Cd) is a widespread, naturally occurring element present in soil, but it is a non-essential element for plant. Many plants can accumulate relatively high levels of Cd, without adverse effects on growth (Kuboi et al. 1986). Cd can be retained for many years in the human body, so the consumption of foods containing high Cd may induce chronic toxicity (FAO/WHO 1995). Therefore, Cd contamination of agricultural soil is a worldwide concern due to the food safety issues and potential health risks.

Despite Cd can enter agricultural soil through various pathways, Cd is usually thought of as being added mainly via phosphate fertilizer and sewage sludge because they contain relatively high concentrations of Cd, e.g. the average value of 54 mg/kg dry matter (DM) for phosphate fertilizer (Lugon-Moulin et al. 2006) and 22.8 mg/kg DM for sewage sludge (Wang 1997) from a small worldwide survey. In contrast, according to some international reports, Cd concentrations in animal manure were very low (below 1 mg/kg DM, Nicholson et al. 1999, Sager 2007) and thus the application of them hardly poses a potential risk for soil and crop (Lipoth and Schoenau 2007, Benke et al. 2008). However, the long-term application of animal manure has attracted our attention, because some abnormally high values of Cd were observed in Chinese animal manure. For example, a large scale investigation in 14 Chinese provinces showed that the highest values of Cd in chicken manure, pig manure (PM), cattle manure and sheep manure were 23.1, 42.7, 51.5 and 4.7 mg/kg DM, respectively (Liu et al. 2005). Consequently, it is necessary to evaluate the potential risk of Cd for soil and crop as a result of the long-term application of animal manure containing high Cd concentration in order to ensure the reasonable use of animal manure in Chinese agriculture.

In this study, we collected soil and crop samples from a long-term experiment (10 years) to assess whether the long-term application of PM would led to a potential risk of Cd for soil and crop. In addition, we calculated the balance of Cd in this

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agricultural system, and offer valid and verifiable scientific data on the optimal use of PM to agricultural soil in the Northeast China.

MATERIAL AND METHODS

Study site and sampling. A long-term fertilization experiment was conducted since 2002 at the Shenyang Ecological Experimental Station of the Institute of Applied Ecology, Chinese Academy of Sciences (41°32'N, 123°23'E). Four treatments were selected in this study: no fertilizer control (CK); low PM rate (M_L); middle PM rate (M_M); and high PM rate (M_H); which were equivalent to 0, 100, 250 and 500 kg total N/ha/year from 2002–2008, and 0, 10, 25 and 50 t fresh weight/ha/year from 2009–2011, respectively. The characteristics of PM are shown in Table 1. The experiment was a two-factorial complete-block design with a split plot arrangement of four PM fertilization rates as main plots and cropping systems as subplots; each plot was replicated three times. The overall subplot size was 9 × 6 = 54 m^2; by considering three replicates, the overall size of each plot was 162 m^2. A three-course rotation with soybean (*Glycine max* L.)-maize (*Zea mays* L.)-maize was conducted in each plot. A more complete description of this experiment field can be provided by Xu et al. (2013).

Crop and topsoil samples (0–15 cm) were collected every year after harvest since 2002. Subsoil (15–30 cm) samples was only collected in 2002 and 2011. Manure samples were collected prior to application. All samples were dried, ground and passed through a 0.15 mm sieve.

Sample analysis. Manure, soil and crop samples were digested using HNO_3-$HClO_4$ (4:1), HF-HNO_3-$HClO_4$ (2:4:1) and HNO_3-$HClO_4$ (4:1), respectively (Lu 2000). The concentration of Cd was measured by a graphite furnace atomic absorption spectrophotometry (GFAAS, PerkinElmer, AA700, Norwalk, USA). Reagent blanks and standard reference samples (GBW10036 for maize and CDGK-SQCI-001 for soil) were also analyzed to monitor analytical accuracy and precision. Three replicates per sample were analyzed.

Data calculation. The loading rate of Cd via PM application (g/ha/year) was estimated by the ratio between the total amount of Cd applied during the experiment (g/ha) and the number of years of the experiment (10 years).

The accumulation rate (μg/kg/year or g/ha/year) was estimated by the ratio between the change of soil total Cd concentration in plots (μg/kg or g/ha) and the number of years of the experiment (10 years).

The output rate through plant removal (g/ha/year) was estimated by the ratio between the total uptake amount of Cd by plant during the experiment (g/ha) and the number of years of the experiment (5 years).

According to some previous papers (Moolenaar and Lexmond 1998, 1999), the elemental field balance in soil system could be identified according to:

$$\Delta soil = \Sigma inputs - \Sigma outputs \quad (1)$$

The main outputs of Cd are represented by surface run-off, crop removal and leaching. However, the output of Cd by surface run-off could be neglected because there were not drains in these flat and isolated plots. Because there were no inputs by irrigation water, animal manure and sewage sludge, the main source of Cd input to CK plot was atmospheric deposition. Therefore, based on a mass balance approach in CK plot ($\Delta soil_{CK}$ = atmospheric deposition – crop removal – leaching), atmospheric deposition rate of Cd was calculated indirectly as:

$$\text{atmospheric deposition (g/ha/year)} = \Delta soil_{CK}\ \text{(0–30 cm)} + \text{crop removal} + \text{leaching} \quad (2)$$

Table 1. The characteristics of pig manure

Characteristic	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011
DM (%)	61.43	58.71	69.53	50.22	53.44	43.61	63.87	61.83	52.46	43.99
C (% DM)	22.94	23.56	22.03	17.52	24.07	25.15	11.18	17.96	16.83	26.66
N (g/kg DM)	11.15	8.77	11.79	16.03	15.85	13.33	10.07	19.25	20.07	23.00
P (g/kg DM)	7.78	8.52	10.05	7.56	9.40	11.39	10.50	17.49	18.59	21.30
K (g/kg DM)	14.00	13.89	17.28	12.29	15.04	15.51	11.02	12.26	15.17	13.48

DM – dry matter

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Table 2. Concentrations of cadmium (Cd) in pig manure and its amount

Treatment	Amount applied (g/ha)										
	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	total
M_L	32.53	71.30	89.84	9.85	10.10	5.41	25.48	12.64	3.32	2.82	263.29
M_M	81.32	177.59	224.60	24.61	25.26	13.51	63.65	31.60	8.29	7.06	657.49
M_H	162.63	355.17	449.19	49.23	50.53	27.02	127.33	63.21	16.58	14.12	1315.01
	concentrations of Cd in pig manure (mg/kg DM)										median
	3.25	12.37	21.02	3.09	2.65	1.08	4.26	2.05	0.63	0.64	2.87

M_L – low; M_M – middle; M_H – high pig manure rate; DM – dry matter

Where the leaching loss from the 30 cm depth in CK plot was obtained from the previous published report (0.15 g/ha/year, Wu et al. 1998).

Based on the calculated atmospheric deposition rate from Eq. (2), the leaching loss from the 30 cm depth in PM plots was calculated indirectly as:

$$\text{leaching (g/ha/year)} = \text{pig manure} + \text{atmospheric deposition} - \text{crop removal} - \Delta\text{soil}_{PM}(0\text{–}30\text{ cm}) \quad (3)$$

Statistical analysis. Difference in Cd concentrations between plant tissues and soil samples collected from different treatments was detected using one-way ANOVA at the 0.05 level with the SPSS 13.0 software (SPSS Inc., Chicago, USA). Relationships between parameters were determined using the Spearman method.

RESULTS AND DISCUSSION

Cd concentrations in pig manure and its input rate. The concentrations of Cd in PM clearly featured a large range from 0.63–21.20 mg/kg DM, with a median value of 2.87 mg/kg (Table 2). Most of these samples exceeded the limits of Cd (1.5 mg/kg DM), in reference to the limit of manure compost in Germany (Verdonck 1997) since there are no available standards in China. These concentrations of Cd are far higher than those reported in Austria (0.02–0.93 mg/kg DM, Sager 2007), England and Wales (0.19–0.53 mg/kg DM, Nicholson et al. 1999). Similar values were also reported in other cities or regions of China (Li et al. 2010, Zhang et al. 2012). Such high concentration of Cd could be due to excessive additives of mineral supplements in animal feeds in China, such as phosphate, zinc sulfate and Zn oxide, which contain high concentrations of Cd (Nong 2002, Zhong and Jiang 2005).

During 10 years, 263.29, 657.49 and 1315.01 g/ha of Cd was applied into the M_L, M_M and M_H plots, respectively (Table 2). The loading rate of Cd were even higher than those loading rates through the application of phosphate fertilizer (17.5 g/ha/year, Schipper et al. 2011) and sewage sludge (30 g/ha/year, Baize 2009). To reduce the long-term pollution risk of hazardous elements in sewage sludge, the Council Directive 86/278/EEC recommended the upper loading rate of 100 g/ha/year of Cd to farmlands. The loading rates of Cd at high PM application rates (131.5 g/ha/year) exceeded this limit, suggesting that the long-term application of PM at high rates could lead to a potential risk for this field.

Cd concentrations in soil and its accumulation rate. Cd gradually enriched in the topsoil (0–15 cm) during long-term application of PM compared to the CK plot (Figure 1). By 2011, Cd concentrations in the topsoil of M_L, M_M and M_H plots increased by 84.34, 183.48 and 362.66%, respectively, but these values did not exceed the maximum allowable concentration (MAC, 1.0 mg/kg

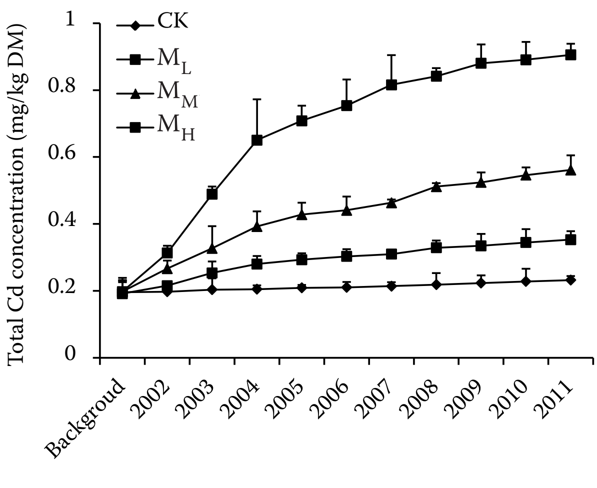

Figure 1. The variation of total cadmium (Cd) concentrations in the topsoil (0–15 cm) during 10 years of pig manure application. CK – no fertilizer control; M_L – low; M_M – middle; M_H – high pig manure rate; DM – dry matter

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Table 3. Change of total cadmium concentration (μg/kg DM) in soil

Treatment	Depth (cm)	2002	2011	Accumulation rate (μg/kg/year)	Accumulation rate[a] (g/ha/year)
CK	0–15	196.0 ± 10.2[a]	232.0 ± 11.7[d]	3.60	6.20
M_L		191.6 ± 9.7[a]	353.1 ± 18.3[c]	16.15	27.86
M_M		198.0 ± 7.1[a]	561.3 ± 20.9[b]	36.33	62.67
M_H		197.8 ± 7.9[a]	905.3 ± 34.7[a]	70.75	122.00
CK	15–30	85.0 ± 6.0[a]	100.4 ± 4.6[d]	1.54	2.89
M_L		84.1 ± 3.1[a]	116.5 ± 8.1[c]	3.24	6.08
M_M		86.4 ± 6.3[a]	138.1 ± 7.6[b]	5.17	9.70
M_H		83.7 ± 4.5[a]	162.0 ± 6.8[a]	7.83	14.68

[a]The calculation assume soil density of 1.15 and 1.25 g/cm^3 for the topsoil and subsoil, respectively. CK – no fertilizer control; M_L – low; M_M – middle; M_H – high pig manure rate; DM – dry matter

DM) in Chinese agricultural soils (GB15168, 1995). This result was similar to previous findings of Han et al. (2008) and Wu et al. (2012), who also reported significantly increasing concentrations of Cd in the topsoil with the application of PM. It should be noted that a significant increase of Cd was also observed in the subsoil (15–30 cm) (Table 3), which is in contrast to the earlier study of Agbenin (2006) and Han et al. (2008), who did not detect a downward movement of Cd after long-term animal manure applications. This discrepancy could be attributed to the differences in soil properties, especially dissolved organic carbon (DOC) and pH.

A small but significant increase of Cd from 196.0–232.0 μg/kg was observed in the topsoil of CK plot (Table 3), which might result from atmospheric deposition due to no other inputs of Cd to this plot. Based on the Eq. 2, atmospheric deposition rate of Cd in this field was estimated to be 10.27 g/ha/year (Table 4). This value was generally higher than those previously measured in different sites around the world, such as England and Wales (1.9 g/ha/year, Alloway 1999) and New Zealand (0.20 g/ha/year, Gray et al. 2003), while it was comparable to that observed in the ChanC9 area of Varanasi (Indian) suffering with heavy traffic load (22.5 g/ha/year, Sharma et al. 2008). Previous studies have also indicated that road traffic emissions were a main source of Cd in atmospheric deposition along the roadside (Sternbeck et al. 2002, Legret and Pagotto 2006). Thus, we concluded that such high atmospheric deposition rate of Cd in this field might result from the road traffic emissions because the experiment field is in close proximity to a highway (approximately 100 m).

The accumulation rate of Cd in the topsoil (0–15 cm) of four treatments ranged from 3.60–70.75 μg/kg/year, and was equivalent to an increase of 6.20–122.00 g/ha/year (Table 3). Based on the accumulation rate of Cd and MAC (1 mg/kg), it was calculated to be 213.6, 40.1, 12.1, and 1.3 years to reach the limit in CK, M_L, M_M, and M_H plots, respectively.

Cd concentrations in crops and its output rate. The concentrations of Cd in maize and soybean were significantly different among four treatments and increased with increasing PM application rates within each year (Table 5). The concentrations of Cd were also significantly correlated between paired soil and crop samples ($r = 0.986$, $P < 0.01$ for stalk of maize; $r = 0.886$, $P < 0.05$ for grain of maize; $r = 0.978$, $P < 0.01$ for stalk of soybean; $r = 0.896$, $P < 0.05$ for grains of soybean). In addition, soybean showed a higher accumulation capacity of Cd than maize, which was consist with previous

Table 4. The input and output rates of cadmium (Cd) during 10 years of pig manure application

Treatment	Atmospheric deposition	Pig manure	Crops removal	Leaching loss
	(g/ha/year)			
CK	10.27	0	1.03	0.15
M_L	10.27	26.33	2.32	0.34
M_M	10.27	65.75	3.12	0.53
M_H	10.27	131.50	4.36	0.73

CK – no fertilizer control; M_L – low; M_M – middle; M_H – high pig manure rate

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Table 5. Cadmium (Cd) concentrations (μg/kg dry matter) in maize and soybean in 2004, 2005, 2009 to 2011

Year	Treatment	Maize		Soybean	
		stalk	grain	stalk	grain
2004	CK	129.4 ± 8.3^{ca}	20.0 ± 1.2^{d}	206.0 ± 10.3^{d}	172.0 ± 3.6^{d}
	M_L	236.4 ± 11.3^{bc}	35.4 ± 1.6^{c}	271.7 ± 9.4^{c}	241.4 ± 5.8^{c}
	M_M	269.9 ± 4.5^{b}	53.1 ± 2.6^{b}	329.6 ± 14.5^{b}	292.5 ± 11.5^{b}
	M_H	358.6 ± 17.4^{a}	62.4 ± 2.4^{a}	424.9 ± 19.7^{a}	404.5 ± 18.7^{a}
2005	CK	137.1 ± 6.1^{c}	21.1 ± 1.1^{d}	208.1 ± 8.6^{d}	188.7 ± 7.7^{c}
	M_L	231.6 ± 10.9^{bc}	30.1 ± 1.3^{c}	279.7 ± 8.6^{c}	274.3 ± 11.1^{b}
	M_M	279.9 ± 12.7^{b}	60.5 ± 2.9^{b}	347.3 ± 12.3^{b}	288.8 ± 15.3^{b}
	M_H	351.3 ± 9.4^{a}	73.3 ± 3.7^{a}	371.7 ± 11.5^{a}	380.4 ± 20.4^{a}
2009	CK	271.5 ± 4.6^{d}	30.0 ± 2.1^{c}	239.4 ± 11.3^{c}	206.1 ± 10.4^{c}
	M_L	311.4 ± 16.2^{c}	45.9 ± 2.2^{b}	325.3 ± 14.6^{b}	308.6 ± 12.2^{b}
	M_M	445.8 ± 21.1^{b}	62.6 ± 2.1^{ab}	389.7 ± 20.0^{a}	483.5 ± 15.6^{a}
	M_H	538.4 ± 24.5^{a}	77.8 ± 3.5^{a}	412.8 ± 23.3^{a}	508.7 ± 23.8^{a}
2010	CK	173.6 ± 8.7^{d}	34.2 ± 1.4^{d}	193.2 ± 8.7^{d}	201.9 ± 9.9^{c}
	M_L	262.0 ± 11.3^{c}	55.7 ± 1.8^{c}	303.4 ± 10.5^{c}	330.5 ± 7.8^{b}
	M_M	407.4 ± 17.4^{b}	72.6 ± 2.4^{b}	390.7 ± 12.4^{b}	376.3 ± 11.5^{ab}
	M_H	542.3 ± 25.6^{a}	82.5 ± 3.7^{a}	437.2 ± 20.5^{a}	401.8 ± 14.6^{a}
2011	CK	122.4 ± 7.3^{d}	23.7 ± 1.6^{c}	203.3 ± 7.2^{c}	204.7 ± 5.6^{d}
	M_L	280.7 ± 10.5^{c}	65.8 ± 2.5^{b}	397.5 ± 10.4^{b}	238.6 ± 12.2^{c}
	M_M	488.5 ± 14.4^{b}	103.7 ± 4.8^{a}	389.1 ± 15.5^{b}	317.6 ± 13.3^{b}
	M_H	529.4 ± 26.0^{a}	128.4 ± 4.7^{a}	449.7 ± 21.3^{a}	439.7 ± 18.1^{a}

Values within each year followed by different letters indicate significant differences at $P < 0.05$ level, determined by the *LSD* test ($n = 3$). CK – no fertilizer control; M_L – low; M_M – middle; M_H – high pig manure rate

studies (Grant et al. 1997, Murakami et al. 2007). More seriously, the Cd concentrations in grains of soybean from all manured plots exceeded the limit (0.2 mg/kg DM), in reference to the Chinese National Food Quality Standard (GB 2762, 2005).

Although the removal rates of Cd through crops also increased with increasing PM application rates (1.03–4.36 g/ha/year, Table 4), this amount was relatively small in comparison to the quantity of Cd supplied by PM application (< 8.49%), suggesting that most of the Cd applied through PM remained in soil after harvest. Also, Čásová et al. (2009) reported that Cd removal by crops represents only small portions (approximately 3.5%) of the total amount added by sewage sludge.

It should be noticed that the input rates of Cd through PM application and atmospheric deposition exceed the sum of its accumulation rate and crop removal rate in all manured plots (Table 4), indicating that Cd leaching occurred in these plots. Based on the Eq. 3, the leaching loss rate from the 30 cm depth of M_L, M_M and M_H plots were calculated to be 0.34, 0.53, and 0.73 g/ha/year, respectively (Table 4), indicating that the PM application enhanced Cd leaching loss. However, these values were lower than the corresponding input rates, suggesting that the Cd derived from PM spreading and atmospheric deposition was almost stored in the 0–30 cm soil layer, and it might not pose a potential risk to groundwater.

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